4. Toxic Contaminants

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OVERVIEW

As our society became more industrialized over the last century, we came to rely on a number of chemicals, some naturally occurring and others newly synthesized, for various industrial and agricultural purposes. These chemicals, and others that occur in a variety of waste streams (e.g., exhaust from internal combustion engines, air pollution from coal burned to generate electricity, waste waters from numerous industrial operations), are released to the environment, sometimes intentionally and sometimes inadvertently or accidentally. With time it has become clear that many of these chemicals are very persistent with extremely low rates of decomposition in the environment, allowing them to linger in sediments and water and accumulate in food webs. It has also become clear, first with DDT[1] and then with other compounds, that some of these compounds have disastrous effects on living organisms.

Concentrations of many contaminants increase as contaminants pass through food webs. A consumer at one trophic level may consume, and accumulate contaminants from, a relatively large number of organisms at lower trophic levels. Organisms at the top of their food webs have the greatest risk of adverse effects since they accumulate the highest concentrations of toxic contaminants in their tissue.

Since DDT in bald eagles was found in the early 1970s to result in thin eggshells and lowered productivity rates, the study of toxic contaminants in the ecosystem has received increasing attention both within the science community and the general public (see a recent report from People for Puget Sound: Schmidt and Johnson 2001).

[1] A number of chemical abbreviations, or acronyms, are used throughout this chapter. An acronym list is given in the Resources section at the end of the report.

Definitions

Bioaccumulation: general term referring to the increase in the concentration of a chemical in a biological organism over time relative to the concentration in the environment.

Bioconcentration: a specific bioaccumulation process by which the concentration of a chemical in an organism becomes higher than its concentration in the air or water around the organism; for example, the concentration of chemicals by filter-feeding bivalves or by fish (through gills) relative to the surrounding water.

Biomagnification: a specific bioaccumulation process by which the concentration of a chemical in an organism reaches higher levels than are found in its food. This leads to higher concentrations of contaminants at higher trophic levels of a food chain.

Persistent, bioaccumulative and toxic pollutants (PBTs): long-lasting substances that can build up in the food chain to levels that are harmful to human and ecosystem health.

This chapter discusses results from recent monitoring activities focusing on a wide range of toxic contaminants in Puget Sound sediments and water as well as shellfish, fish, birds and mammals. The new findings and accomplishments in this chapter include the following highlights.

Sediment

Comparing surface sediment samples collected in 2000 to results from 1989 through 1996 at 10 long-term Puget Sound sites showed:

- Decreases in metal concentrations, particularly mercury and copper
- Increases in polycyclic aromatic hydrocarbon (PAH) concentrations
- Substantial increase in benzoic acid at all sites
- Particularly high PAH concentrations at the Thea Foss Waterway (Commencement Bay) relative to the other sites
- Particularly high metal concentrations at Sinclair Inlet relative to the other sites.

Analysis of sediment samples from 300 randomized sites collected in 1997 through 1999 in Puget Sound showed:

- The majority of observed sediment contamination was located in urban waters including Bellingham Bay, around March Point, Sinclair Inlet, Everett Harbor, Elliott Bay and Commencement Bay.
- Greatest toxicity was found in Everett Harbor, Elliott Bay, Commencement Bay and the Port of Olympia based on a series of four toxicity tests designed to gauge impacts on biota.
- Greatest degree of degraded sediments was found in central Puget Sound, followed by Whidbey Basin, south Puget Sound and Hood Canal regions, based on the weight of evidence developed through a triad of sediment quality information.
- A portion of each study region, including the majority of Hood Canal, showed no signs of sediment degradation.

Shellfish

- Dungeness crab in Puget Sound accumulate PAHs, with the greatest exposure occurring in crab in urban areas, suggesting that they are suitable as indicator species to quantify PAH exposure in marine biota.
- Analysis of butter clams from six sites in central Puget Sound showed metal concentrations below Food and Drug Administration (FDA) standards and no detectable organic compounds except benzoic acid, which was prevalent in all samples.

Fish

- Polychlorinated biphenyl (PCB) concentrations in Pacific herring were higher in fish from central and southern Puget Sound relative to fish from the northern Sound and the Strait of Georgia. These PCB concentrations were generally below values suggested by the National Marine Fisheries Service (NMFS) as thresholds for possible adverse effects in salmonids, although there is a potential for adverse effects in herring stocks in central Puget Sound.

- At long-term sampling stations, English sole in two urban bays (Elliott and Commencement) were observed to have significantly higher risk of liver disease than the intermediate risk measured at Sinclair Inlet and Port Gardner and the lower risk observed in the Strait of Georgia and Hood Canal.
- Monitoring of English sole, rock sole and starry flounder at a site where highly contaminated sediment was capped with clean sediment, showed reduced risk of liver disease associated with reduced exposure to PAHs.

Birds and Mammals

- A study of PCB contaminants in orcas based on samples from 1993 through 1996 showed very high levels of contamination with clear relationships to food source. Transient orcas that feed from higher trophic levels (marine mammals) were the most highly contaminated of the whales studied. Southern residents were four to six times more contaminated than northern residents presumably because their food source comes from more contaminated areas of Puget Sound and the Strait of Georgia.
- A study of contaminants in Hood Canal bald eagles concluded that while the population was growing, PCBs were negatively affecting the eagles and causing lower productivity rates relative to populations elsewhere in Washington State.

Clopyralid: A new concern

As new compounds are regularly synthesized by the chemical industry, there is an ongoing need to monitor for these compounds in the environment and their toxicity to living organisms. A recent example is the compound clopyralid, which is not currently known to have impact on the marine environment but has introduced a serious problem to the compost industry. Clopyralid is an herbicide produced by Dow Agrosciences that is persistent and toxic to some plants at very low concentrations. The compound was introduced into commercial compost through trimmings and agricultural waste and was later found to be responsible for killing plants in gardens and nurseries where the compost was used. This problem, first observed in Spokane (Bezdicek et al. 2000), is the subject of intense debate and sends a clear message about the need for the continuing study of toxic compounds in the environment.

FINDINGS

Sediment Contamination

All sediments in Puget Sound (and areas of net deposition out to the ocean) have concentrations of chemicals that are elevated over pre-industrial levels as a result of human activities. The pattern of this elevated concentration has two components: discrete hot spots around sources and a general smog representing contamination carried far from sources by currents or as airborne contaminants.

There are many different tools for measuring levels of contamination and biological harm in sediments. The State of Washington has adopted sediment management standards that use a combination of tests to pass or fail sediments for the purposes of state sediment management. Because animals have access to buried contamination, the state applies the sediment standards to samples from the top 10 cm to determine level of degradation for regulatory purposes.

The PSAMP sediment monitoring program samples recently deposited sediments (0-3 cm) to better focus on trends in contamination. Past studies have shown the rates of change in sediment contaminant concentration to be generally low in Puget Sound. By limiting the sampling to recent sediments only, PSAMP is more likely to reveal temporal patterns rather than lumping the record of deposited sediment represented by a 10 cm sample.

PSAMP scientists in the Washington State Department of Ecology use the state sediment management standards to evaluate measurements from these 0-3 cm samples, but it is important to note that the results are not meant to be used for regulatory purposes (which require application of standards to 0-10 cm samples). PSAMP results may differ from analysis of 0-10 cm samples that would be used for regulatory purposes. Furthermore, PSAMP scientists have assessed degraded sediments based on their analysis, and this is distinct from the legal definition of contaminated sediments used for regulatory purposes in Washington State.

Long-term Trends in Sediment Condition

As one element of the PSAMP sediment component, Department of Ecology scientists collected surficial sediments in 2000 from 10 of the original PSAMP sediment component sampling stations (Llansó et al. 1998a, 1998b; PSWQAT 1998; see Figure 4-1) and analyzed these samples for 121 toxic chemicals (including metals, PAHs, PCBs, pesticides, and other organic contaminants). The 10 stations are located in or near the Strait of Georgia, Bellingham Bay, North Hood Canal, Port Gardner, Shilshole, Sinclair Inlet, Point Pully, Thea Foss Waterway, East Anderson Island, and Budd Inlet and are sampled at five-year intervals. These stations were chosen for their widespread geographic distribution throughout the Sound, their co-occurrence with other PSAMP component (i.e., fish, water column) sampling locations, the pre-existence of long-term (30-year) data sets for some locations, and for the occurrence of differing benthic community assemblages at each location. Results for 2000 were summarized and compared with the historical PSAMP (1989-1996) data set to determine whether any significant changes occurred in the levels of chemical contaminants at these 10 "sentinel" PSAMP sediment stations over time.

Overall, target compound concentrations were above detection limits in 32.5 percent of the analyses conducted on sediment samples collected in 2000, similar to that for the data from 1989 through 1996 (32.2 percent). Sediment quality problems, represented by concentrations above state sediment quality standards, observed in 2000 were generally consistent with the evidence from previous years (e.g., mercury contamination in Sinclair Inlet and PAH contamination in the Thea Foss Waterway). But the 2000 results also indicated a problem level of benzoic acid in inner Budd Inlet. Results were compared to state sediment quality standards (SQS and CSL values—Washington State Sediment Management Standards, Chapter 173 204WAC) and sediment guidelines (ERM values—Long et al. 1995) derived from data collected nationally.

Sediment contaminant levels at each station were compared over time for the 53 compounds that were measured at detectable levels. Data that qualified as "undetected" (i.e., measured in quantities at or below the analytical laboratory equipment's ability to detect a compound) were excluded from these analyses. Significant increase or decrease in sediment contaminant levels between the historical and the most recent samples was determined for each compound by comparing the median and 95 percent confidence limits for the data from 1989 through 1996 with the value measured in 2000 at each station. In addition, analyses were conducted to determine, for each compound, the predominant change in concentration value (i.e., increasing, decreasing, no change) for all 10 stations combined. These changes are described, in part, below.

- Of the 121 compounds reexamined in 2000, 68 were undetected at the quantitation limits of the laboratory instrumentation.
- Measurements for 37 of the 53 detected compounds in 2000 were statistically significantly different from the median values from 1989-1996 at one or more of the 10 locations sampled.
- Overall, there were more increases (75) than decreases (52) in the levels of individual compounds at individual stations (530 possible). Closer examination showed that metals concentrations

Figure 4-1. PSAMP long-term sediment monitoring stations.

Source: Washington State Department of Ecology

decreased substantially (41 decreases versus five increases), while PAHs increased in similar proportion (45 increases versus nine decreases). Other organic compounds accounted for 26 increases and four decreases.

- Point Pully (station 38) sediments were remarkable in that the levels of 17 compounds (10 metals, six PAHs, and one other organic, β-coprostanol) decreased, while only three organic compounds (benzoic acid, cholesterol, and cymene) increased.

Benzoic Acid

Benzoic acid is a naturally occurring compound in many plants and animals, such as berries and shellfish. This compound is the degradation product of hippuric acid, present in the urine of herbivores and also, to a small extent, in humans (Wibbertmann et al. 2000). It is always found in tissues of marine bivalves and other marine herbivores, as it is part of the metabolic pathway for these organisms. Anthropogenic sources of benzoic acid into the aquatic environment are primarily from its use as an anti-microbial food preservative. Benzoic acid is also produced as an intermediary byproduct in the reduction of aromatic compounds, such as phenol and cresol (dimethyl phenol).

Benzoic acid toxicity to humans is low, although it can cause allergic reactions in some people. Benzoic acid is thought to exhibit low toxicity to various aquatic organisms. Most toxicity studies have been conducted with freshwater organisms, and the toxicity to marine organisms is not well known. Benzoic acid is rapidly biodegradable, has low bioaccumulation potential, is rapidly metabolized by biota, and does not adsorb to sediments (Wibbertmann et al. 2000).

Benzoic acid is often measured in sediments when organic pollutants are a concern. Results for this compound are often difficult to interpret as high levels may indicate anthropogenic input but may also indicate an abundance of benthic organisms and a healthy environment. When sampling sites are located in riparian or vegetated areas, high benzoic acid levels may be found due to the nearby plant material. For example, high benzoic acid levels detected at one location in the Duwamish River appear to derive from overhanging blackberry shrubbery along the riverbank (S. Mickelson, pers. comm.). Therefore, when interpreting benzoic acid results it is necessary to evaluate many factors, including site characteristics and benthic community data, to determine if the source of this compound might be natural or anthropogenic.

- The concentrations of eight metals decreased at Budd Inlet (station 49).
- The locations with the most numerous increases in PAH levels were East Anderson Island (station 44), with 11 compounds increasing (none decreasing), Bellingham Bay (station 4), with 8 compounds increasing (none decreasing), and Thea Foss Waterway (Commencement Bay, station 40), with 8 compounds increasing and one decreasing.
- Benzoic acid levels were measured an order of magnitude higher in samples collected in 2000 than they were historically. These results were strongly statistically significant. There was no evidence of an increasing trend in the early 1990s, and no data for the intervening years are available.
- Cholesterol and β-sitosterol levels were both significantly higher in samples collected in 2000 than they were historically at five of the 10 stations. At the Strait of Georgia (station 3), North Hood Canal (station 13R), and at Shilshole (station 29) concentrations of both compounds increased.
- Copper levels were significantly lower in samples collected in 2000 than they were historically at seven of the 10 stations.
- Mercury concentrations were significantly lower in samples collected in 2000 than they were historically at five of the 10 stations.
- Naphthalene and total LPAH were significantly higher in samples collected in 2000 than they were historically at five of the 10 stations.
- Total HPAH levels in 2000 were, on average, 1.5 times higher than they were historically, while total LPAH levels were 2.5 times higher.
- At Thea Foss Waterway (station 40) the LPAH and HPAH concentrations were about 25 times as high as at the other stations.
- Over all years, Sinclair Inlet (station 34) had higher levels of metals, particularly copper, lead, mercury, silver, and zinc, than any of the other locations.

Other trend analyses were conducted on these data that are not reported in this document. A publication describing the full set of PSAMP sediment temporal trend analyses is in preparation (Marine Sediment Monitoring Team, in prep.), and will be released at a later date.

Ecology's 1997-1999 Evaluation of Sediment Quality throughout Puget Sound

As described in the *2000 Puget Sound Update,* Department of Ecology scientists refocused the PSAMP Sediment Component's spatial monitoring element to form a three-year partnership with the National Oceanic and Atmospheric Administration's (NOAA's) National Status and Trends Program. This study used a stratified-random sampling approach to estimate the percentage of the Puget Sound study area in which sediment quality is significantly degraded based on a variety of evaluation approaches.

Three hundred samples of recently deposited sediment were collected during June and July of 1997, 1998, and 1999, at locations extending from the U.S./Canadian border, south to Olympia, and west to Hood Canal. Sediments from each location

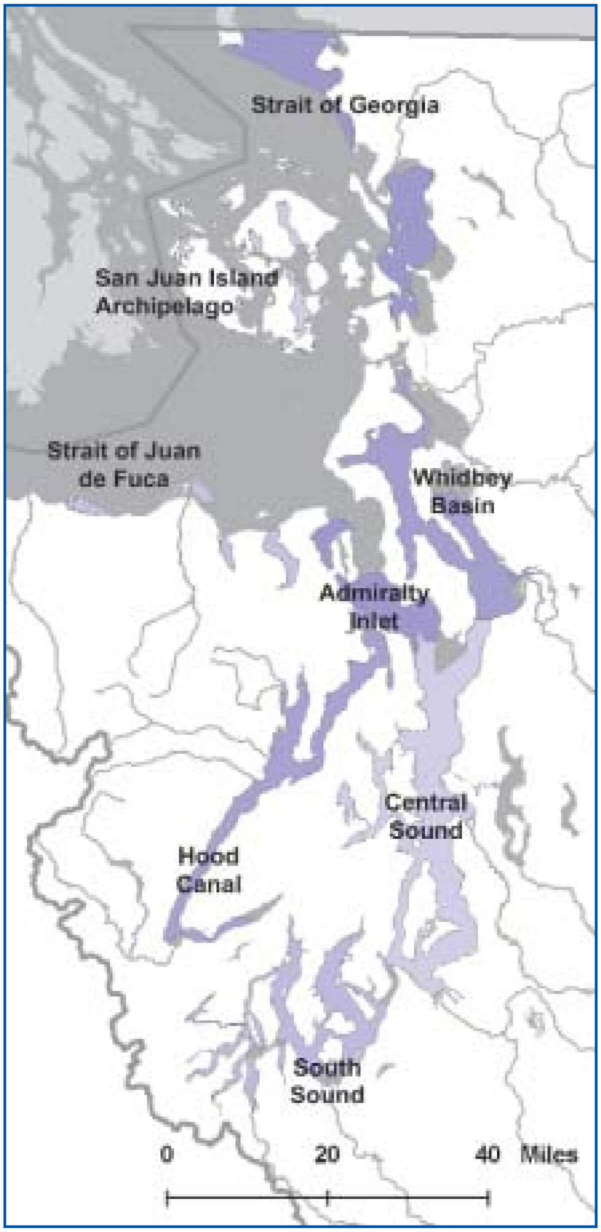


Figure 4-2. The six regions used for the 1997-1999 Ecology-NOAA sediment monitoring study. Two additional regions are shown that are used by the Department of Ecology in continuing PSAMP sediment monitoring—the Strait of Juan de Fuca and San Juan Island Archipelago regions.

Source: Washington State Department of Ecology

were analyzed for more than 160 toxic contaminants and physical sediment characteristics, tested for toxicity in four laboratory tests, and characterized by the benthic infaunal organisms dwelling within them. The results were summarized in three separate reports (Long et al. 1999, 2000, 2002) and characterize six geographic regions including the Strait of Georgia, Whidbey Basin, Admiralty Inlet, central Sound, south Sound, and Hood Canal (Figure 4-2). The distribution and spatial extent of sediment chemical contamination, toxicity, and benthic infaunal distribution was determined for all regions, along with a Sediment Quality Triad "weight-of-evidence" to characterize sediments throughout the Puget Sound study region. The triad includes three tests (sediment contaminants, sediment toxicity, and infaunal invertebrate community structure) that were examined simultaneously at each station.

Distribution and Spatial Extent of Chemical Concentrations in Puget Sound Sediments

The Ecology-NOAA study tested sediments to determine the concentration of more than 160 chemical contaminants.

Contaminants exceeding various state standards in 1997-99 sampling

Arsenic
Copper
Mercury
Lead
Silver
Zinc
Individual LPAHs & HPAHs
Total LPAH
Total HPAH
Total PCB congeners
Total Aroclor
Phthalate esters (3)
Phenol
Phenolic compounds (3)
Chlorinated aromatic compounds (3)
Benzoic Acid
Benzyl Alcohol
Dibenzofuran

Throughout Puget Sound, 22 compounds or compound groups were measured in concentrations exceeding state sediment criteria (Sediment Quality Standards, SQS; or Cleanup Screening Levels, CSL) and/or national sediment guidelines (Effects Range Median, ERM values). These include six priority pollutant metals (arsenic, copper, mercury, lead, silver, and zinc), individual LPAH and HPAH compounds (low and high molecular weight PAHs, respectively) and total LPAH and HPAH sums, total PCB congener and Aroclor sums, three phthalate esters, phenol and three substituted phenolic compounds, two chlorinated aromatic compounds, and three miscellaneous extractable compounds (benzoic acid, benzyl alcohol, and dibenzofuran).

The average concentration, spatial extent of contamination (i.e., the number of stations and proportion of study area represented), and locations in which each of the 22 chemical contaminants or contaminant groups exceed state sediment criteria and/or national sediment guidelines for the different regions examined are summarized in Long et al. (in prep). As expected, the majority of the contaminated locations are urban/industrialized areas, including Bellingham Bay, March Point, Sinclair Inlet, Everett Harbor, Elliott Bay, and Commencement Bay.

The spatial extent of chemical contamination relative to three different guidelines or standards is summarized in Figure 4-3 for the six geographic regions of Puget Sound. The bar charts shown represent the percent of stations in which at least one compound was measured at levels above one or more of the three sediment guidelines and standards, and the proportion of each study region that these stations represent. The central Puget Sound region had the greatest percent of stations exceeding ERM, SQS, and CSL values (located primarily in the urban/industrial embayments of Elliott Bay, Commencement Bay, and Sinclair Inlet). The Whidbey Basin had the next greatest number of ERM and CSL exceedances (with contaminant problems primarily in Everett Harbor). The total percent area of each region contaminated above criteria was largest in the Strait of Georgia for ERM and SQS values, and largest in the Whidbey Basin for CSL values. No chemical contaminant values exceeded guidelines in the Admiralty Inlet region.

Spatial Extent of Toxicity in Puget Sound Sediments

In a second part of the Ecology-NOAA study, scientists subjected sediments from the six Puget Sound study regions to a battery of four toxicity tests that tested different phases of the sediment, different types of organisms, or were sensitive to different suites of contaminants that might be present in the sediments. These tests included the 10-day amphipod survival test conducted with bulk sediment samples on an amphipod, *Ampelisca abdita*. This test is a measure of the survival rate of adult amphipods in bulk sediments. A second test, the sea urchin fertilization test, was conducted with sediment pore water samples on *Strongylocentrotus purpuratus* gametes to test the reproductive sensitivity of echinoderms to the presence of toxicants in the sediments. The third and fourth tests, the microbial bioluminescence (Microtox™) and cytochrome P450 HRGS tests, were conducted with organic solvent extracts of sediment samples. The Microtox™ test is a measure of the toxicity

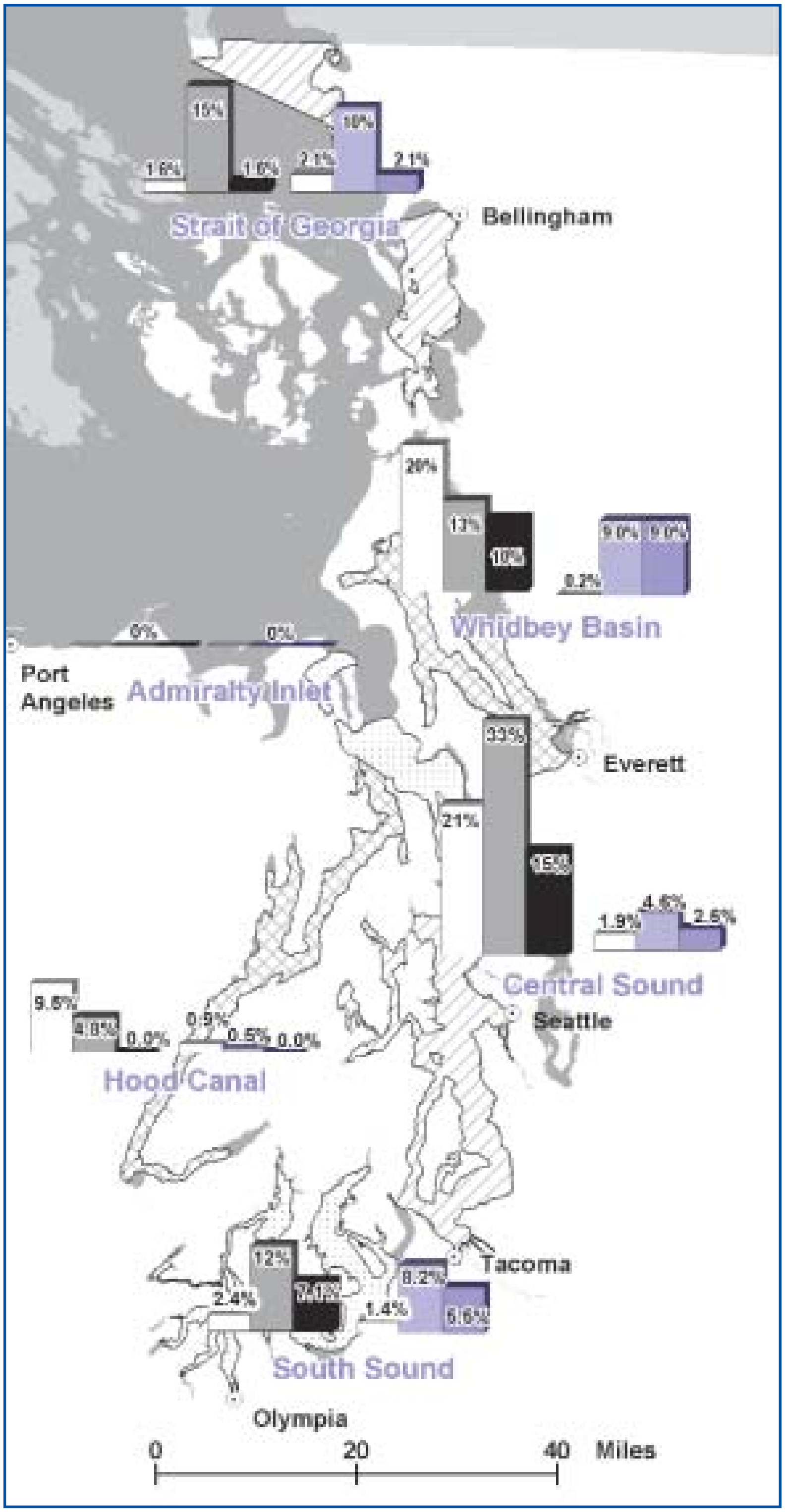
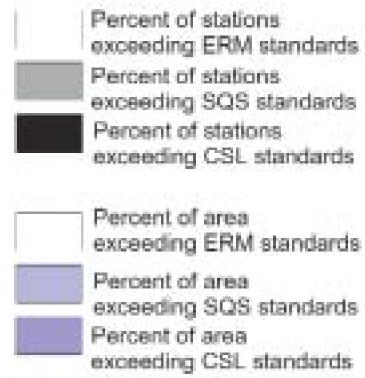


Figure 4-3. Chemical contamination by study region relative to three standards. For each region, one chart shows the proportion of stations exceeding each set of standards for at least one contaminant. The other chart shows an estimate of the proportional area that exceeds the standards.



Source: Washington State Department of Ecology

Figure 4-4. Results of four toxicity tests by study region. For each region, one chart shows the proportion of stations exceeding each test criteria. The other chart shows an estimate of the proportional area that exceeds the test criteria.

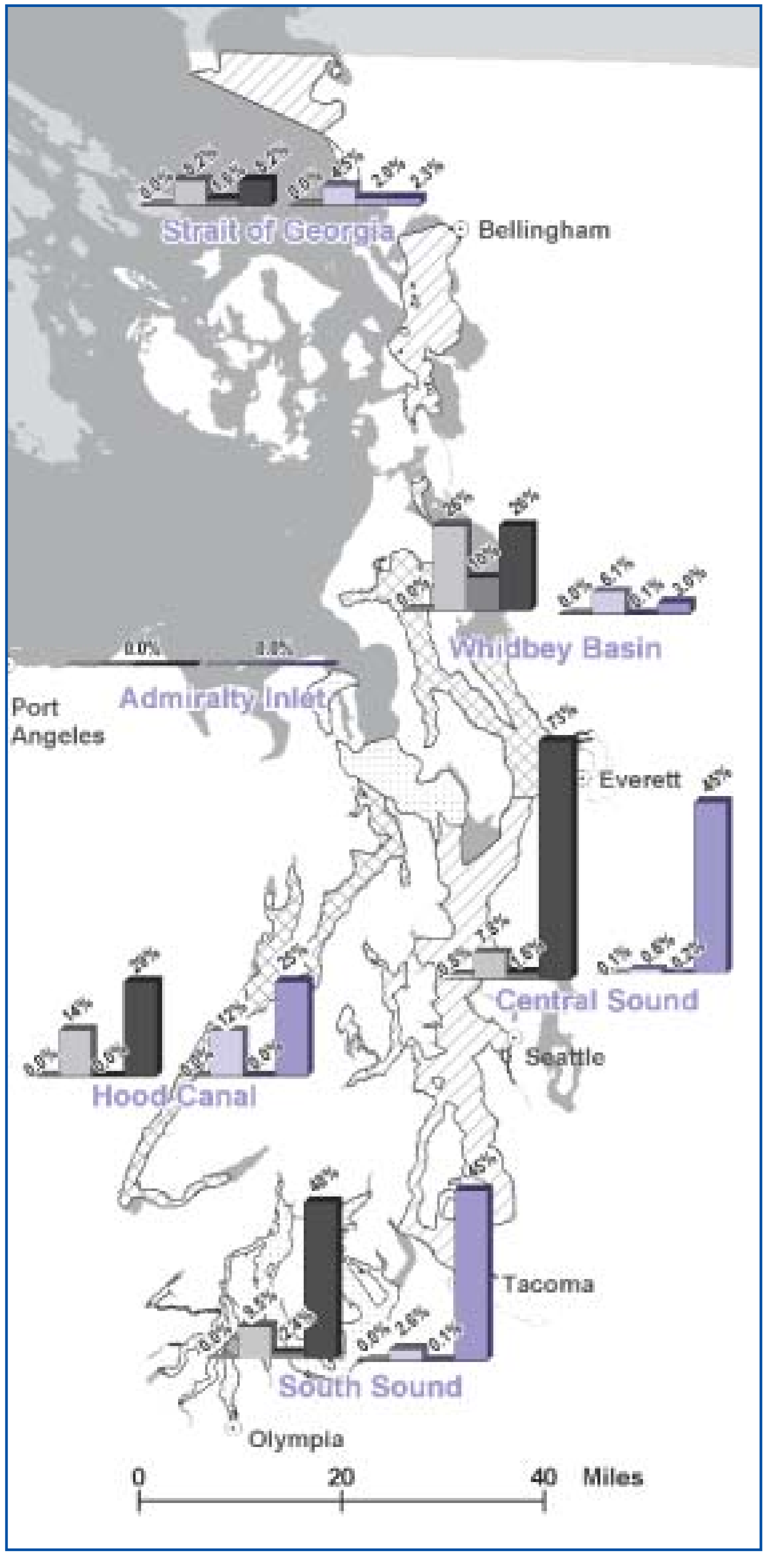
Percent of stations exceeding toxicity test

- Amphipod survival
- Urchin fertilization
- Microbial bioluminescence
- Cytochrome P450 HRGS

Percent of study region area exceeding toxicity test

- Amphipod survival
- Urchin fertilization
- Microbial bioluminescence
- Cytochrome P450 HRGS

Source: Washington State Department of Ecology



of an organic solvent extract of the sediments (i.e., all bioavailable and bound contaminants in the sediments). The cytochrome P450 HRGS assay is an indicator of the presence of PAHs, PCBs, dioxins and furans.

Estimates of the spatial extent of toxicity for the six geographic regions of Puget Sound are summarized in Figure 4-4. The highest levels of toxicity and greatest concurrence among toxicity measures occurred in the urban/industrialized embayments, including Everett Harbor, Elliott Bay, Commencement Bay, and the Port of Olympia, while significant, single test responses occurred in other urban and rural locations. In central and south Sound and Hood Canal, stations displayed the greatest percent of stations and percent area with the cytochrome P450 HRGS assay, an indicator of the presence of PAHs, PCBs, dioxins, and furans, with the majority of these responses occurring in the urban bays of the central Sound region. The urchin fertilization porewater test, representing the reproductive sensitivity of echinoderms to the presence of toxicants in the sediments, displayed the next greatest percent number of stations and area of responses in the Whidbey Basin. Microtox™ test response, representing the toxicity of an organic solvent extract of the sediments (i.e., all bioavailable and bound contaminants in the sediments), was much lower throughout the total study area, the greatest number of responses were in the Whidbey Basin (Everett Harbor). Toxicity was observed in only one of the 300 amphipod tests (Port Washington Narrows) which test the survival rate of adult amphipods in bulk sediments.

Benthic Infaunal Communities

The types and quantities of chemical compounds deposited in the estuarine sediments of Puget Sound play a role in determining the toxicity of these sediments and, ultimately, the composition of the invertebrate communities that can inhabit them. Organisms living within the surface sediments were collected, identified, and counted for each of the 300 stations sampled. Sixty-one of the 300 stations appeared to have impaired invertebrate communities, based on a comparison of benthic indices including total organism abundance, taxa richness, the evenness of the distribution of individuals, and the number and composition of the dominant organisms found at each station (Long et al. 1999, 2000, 2002). Stations that appear to have impaired communities were all found in harbors, embayments, and inlets including Bellingham Bay, Oak Harbor, Everett Harbor, Port Gardner, Sinclair and Dyes Inlets, an inlet off Port Washington Narrows, Elliott Bay and the Duwamish River, Commencement Bay waterways, Gig Harbor, Budd Inlet and the Port of Olympia, Oakland Bay at Shelton, Port Ludlow, and Port Gamble.

Sediment Quality Triad "Weight-of-Evidence"

To determine the overall quality of the recently deposited sediments sampled throughout Puget Sound, all three elements of the Sediment Quality Triad (i.e., sediment contaminant levels above critical values, sediment toxicity, and impaired infaunal invertebrate communities) were examined simultaneously to determine the extent to which they co-occurred at each station. Sediment quality for each station was ranked based on a "weight-of-evidence" approach, and four categories of sediment quality were generated to define each station, based on the sum of the number of impaired parameters measured at each station.

These four categories of sediment quality include:

- **High Quality** (no degradation detected)
- **Intermediate/High Quality** (degradation detected in one test)
- **Intermediate/Degraded Quality** (degradation detected in two tests)
- **Degraded Quality** (degradation detected in three tests)

The percent of stations in each of these four sediment quality categories, and the percent area in each category were summed for each of the six Puget Sound sediment study regions. These spatial extent results are displayed in Figure 4-5. Based on this index, the central Sound region displayed the greatest percent area with degradation in all three sediment tests, followed by the Whidbey Basin, Hood Canal and then the south Sound regions. Hood Canal had the greatest percent area with no degradation detected in the three tests, followed by south Puget Sound, Whidbey Island, Admiralty Inlet, the Strait of Georgia and the central Sound region.

Toxic Contaminants in Water

Contamination of the Sea Surface Microlayer in Burrard Inlet, Vancouver, B.C.
Scientists from British Columbia's Ministry of Environment Land and Parks (now the Ministry of Water, Land and Air Protection) studied contamination in the surface microlayer of Burrard Inlet, an embayment of the Strait of Georgia at Vancouver, in 1999 and 2000 (Moore and Freyman 2001).

Results indicated elevated levels of contaminants in the Burrard Inlet microlayer, at levels similar to those reported elsewhere, e.g., Puget Sound (Batelle 1988) and Chesapeake Bay (PTI 1990). Copper and other metals occur in the microlayer in concentrations up to 20 times the relevant B.C. water quality criteria. Some PAHs, such as benzo(a)pyrene and chrysene, occur at concentrations almost 400 times their criteria.

The observed levels of contamination of the microlayer are significantly greater than the concentrations measured in the underlying surface waters, which have traditionally been monitored to determine water quality conditions. Thus, a lack of microlayer monitoring may downplay the actual impacts of contaminants to water quality and marine life forms.

The limited sampling accomplished by B.C. scientists suggests that microlayer contamination may be restricted to areas immediately surrounding point source discharges or to embayments (adjacent to developed lands) affected by non-point sources of pollution (including atmospheric deposition and runoff). While they do not expect that significant microlayer contamination extends over large areas of Georgia Strait, the B.C. scientists recommend further monitoring to accurately characterize the extent and environmental significance of microlayer contamination.

Surface Microlayer

The surface microlayer is approximately 50 microns thick. It has significant value for the eggs and larvae of numerous marine organisms utilize this area as a "nursery". However, the same factors that allow eggs and larvae to collect in this region also allow contaminants to be concentrated at this interface between the marine water and the atmosphere. Contaminants from sources such as stormwater, effluent discharges, spills, and atmospheric deposition can accumulate in the surface microlayer.

The microlayer also has significant contact with habitats in the intertidal zone. As water levels fluctuate, organisms are repeatedly exposed to these elevated concentrations of contaminants.

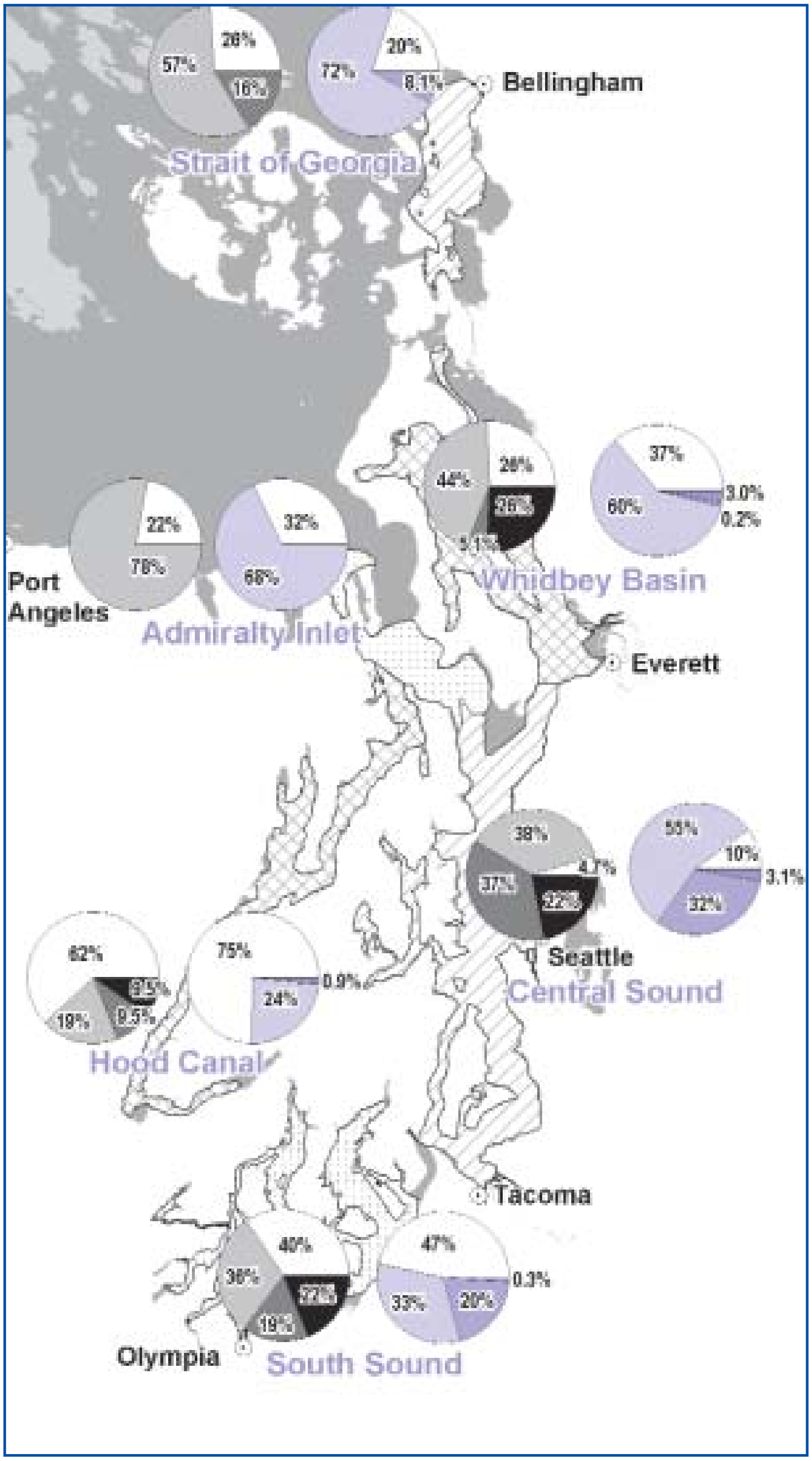
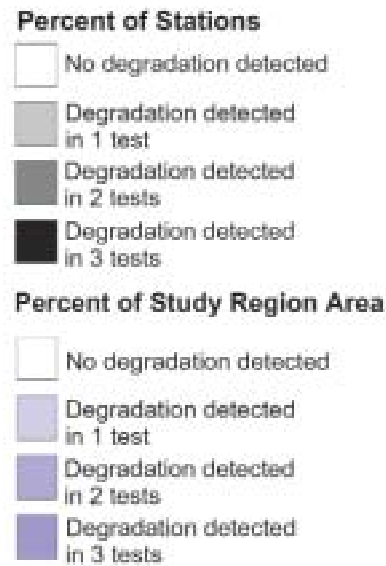


Figure 4-5 Sediment triad results by study region. For each region, one pie chart shows the percentage of the stations in four categories based on results from the three sediment tests. The other pie chart shows an estimate of the proportional area of the region that falls into the four categories.



Source: Washington State Department of Ecology

Metal Contaminants in Shellfish

State and federal criteria do not exist for acceptable levels of metals in shellfish tissues. The U.S. Food and Drug Administration (FDA), however, has established guidance values termed Levels of Concern for mollusks for five metals: arsenic, cadmium, chromium, lead and nickel. Additionally, the FDA has established an Action Level in fish and shellfish tissues of 1.0 mg/kg for mercury. Food products that exceed the Action Level cannot be commercially traded, an important distinction from Levels of Concern.

Toxic Contaminants in Shellfish

Polycyclic Aromatic Hydrocarbons in Dungeness Crab

Washington State Department of Fish and Wildlife scientists, from the PSAMP Fish Component and the Oil Spill Team, conducted a study in the spring of 2001 to evaluate background levels of PAHs in the hepatopancreas of Dungeness crab (*Cancer magister*). The goals of this pilot project were to determine whether crabs are sufficiently exposed to toxics (as measured by tissue burdens) to warrant their use as a monitoring species, especially for natural resource damage assessments in the event of an oil spill. This information is critical in helping the state Department of Fish and Wildlife determine if its trust resources have been injured following a spill. For this study, state Department of Fish and Wildlife scientists were interested in evaluating and refining the procedures for collecting crab tissue samples and to measure PAH exposure of crabs in Puget Sound to use as a baseline from which to compare conditions after an oil spill.

Dungeness crabs are both ecologically and economically important in Puget Sound. Their pelagic larvae are an important prey for out-migrating coho salmon and other fishes, and adult crab are eaten by a wide variety of Puget Sound's predators. In addition, Dungeness crab supports important tribal, commercial and recreational fisheries throughout much of Puget Sound. Because of their benthic feeding mode, abundance, and ubiquity in the food chain, they represent an important potential pathway for toxic contaminants in the ecosystem. In addition to exposure to toxics that predators experience from consuming contaminated benthic crabs, PAHs and other organic toxics such as PCBs may be passed to pelagic larvae from female crabs, resulting in transfer of toxics from sediments to the water column, where salmon, herring and other pelagic species feed.

In this study, PAHs were successfully measured in samples of crab hepatopancreas from four locations in Puget Sound and the observed pattern of contamination in crabs was correlated with degree of PAH sediment contamination. Preliminary analysis of these results shows that Dungeness crab from Commencement Bay (Thea Foss Waterway), one of Puget Sound's more PAH contaminated habitats, had significantly greater total PAH (TPAH) concentration than those from Port Gardner (situated near Everett), Vendovi Island, (outside of Bellingham Bay) and Cherry Point (Figure 4-6).

The crabs' primary detoxifying organ, the hepatopancreas, is less capable than vertebrate liver of metabolizing complex organic toxics like PAHs. For this reason their bodies accumulate PAHs, whereas fish metabolize and excrete them. Hence, crabs are better suited than fish as monitors of PAH accumulation in marine biota. In the future, state Department of Fish and Wildlife scientists will look at the patterns of distribution of individual PAH compounds to help understand better the origins of these toxics. If additional monitoring funds become available, they plan to analyze muscle tissue from these crabs as a first step in the process of assessing risk to humans from consuming this species. Scientists on the Oil Spill Team will also continue to investigate other potential indicator species such as the graceful crab (*Cancer gracilis*), a smaller, non-harvested crab, to characterize baseline or background conditions of oil-based toxic contaminants in Puget Sound biota.

Toxic Contaminants in Shellfish—King County Monitoring

In 1999 and 2000, King County scientists collected butter clams (*Saxidomus gigantea*) from six Puget Sound locations. Stations were located from Richmond Beach near the northern border of King County to Normandy Park in the south (Figure 4-7). Whole clam tissues were analyzed for metals, pesticides, PCBs, and semi-volatile organics.

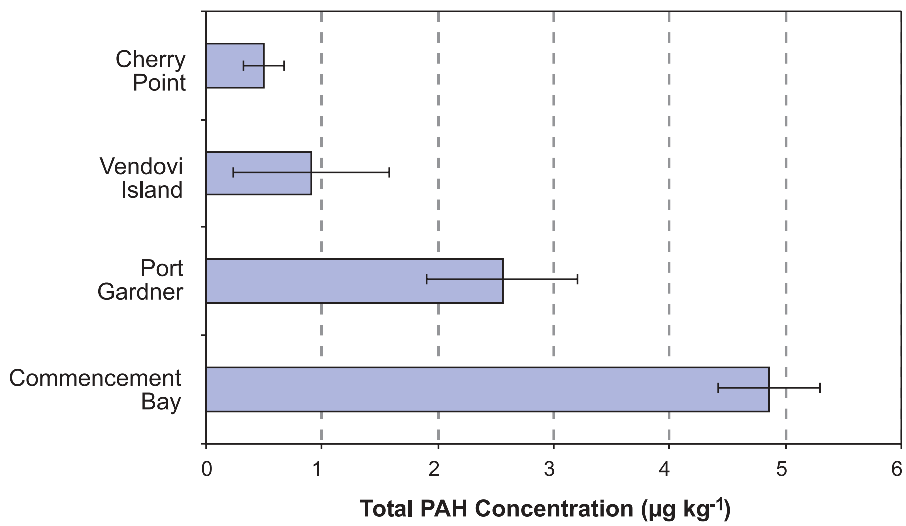


Figure 4-6. Concentration of total PAHs in the hepatopancreas of Dungeness crab from four Puget Sound locations (mean of 3 to 4 replicate composites for each location, 5 crabs per composite, ± std. error). Mean concentration from Commencement Bay significantly greater than the other three stations, no significant difference between Port Gardner, Vendovi Island and Cherry Point, (one-way ANOVA, Tukey's post hoc multiple range comparison, $p<0.001$).

Source: Washington State Department of Fish and Wildlife

Figure 4-7. King County shellfish toxic contaminant monitoring stations.

Source: King County Department of Natural Resources and Parks

Table 4-1. Concentration of metals detected in butter clams in King County marine waters. Beryllium concentrations were measured but were below detection limits for all samples.

Source: King County Department of Natural Resources and Parks.

	Concentration (mg/kg dry weight)									
	Arsenic	**Cadmium**	**Chromium**	**Copper**	**Lead**	**Mercury**	**Nickel**	**Selenium**	**Silver**	**Zinc**
FDA Level of Concern	55	3	11	---	0.80	---	80	---	---	---
1999										
Carkeek Park	12.5	0.257	2.22	8.00	0.951	0.040	4.30	1.46	2.81	69.7
West Point N	16.9	0.302	3.19	9.38	0.780	0.062	4.85	1.69	2.50	65.0
West Point S	15.3	0.291	3.16	9.09	0.575	0.042	5.35	1.77	1.65	66.1
Alki Point	21.7	0.315	2.44	9.42	0.519	0.050	5.74	2.03	2.77	76.7
Richmond Beach	17.5	0.379	2.43	21.4	0.964	0.060	4.51	2.18	3.90	71.9
Normandy Beach	23.4	0.369	2.63	7.70	0.589	0.073	5.11	2.62	5.39	81.0
2000										
Carkeek Park	13.4	0.344	1.88	8.27	0.804	0.041	5.34	2.04	4.34	70.9
West Point N	19.9	0.285	4.66	10.8	0.623	0.069	6.00	3.36	6.57	78.9
West Point S	15.1	0.314	4.64	10.5	0.621	0.035	7.40	2.29	1.00	79.7
Alki Point	18.2	0.296	3.52	9.3	0.507	0.033	5.43	2.02	3.13	77.5
Richmond Beach	15.3	0.466	1.65	7.05	0.801	0.045	5.80	2.24	3.35	78.4
Normandy Beach	18.7	0.306	3.11	8.2	0.625	0.037	4.50	2.19	5.85	76.5

Metals concentrations at all stations were well below the FDA Levels of Concern and mercury was below the Action Level. Beryllium was not detected in any sample either year. Cadmium, chromium, lead, mercury, nickel, selenium, and zinc concentrations varied only slightly between stations and were similar to values detected in previous years (Table 4-1). Normandy Park generally had the highest concentrations detected for most of the metals in 1999. In 2000, the highest metals concentrations were generally detected in sediments collected from West Point. Copper was unusually high at Richmond Beach in 1999, 21.4 mg/kg dry weight, which is nearly twice as high as the highest copper value in previous years.

Only one organic compound was detected in the shellfish samples: benzoic acid. Benzoic acid has always been detected in all shellfish samples. Benzoic acid is used as a food preservative and an anti-fungal agent and is also a degradation product of metabolic processes (see **Benzoic Acid** sidebar in earlier section of this chapter).

NOAA Mussel Watch Results for Puget Sound

Mussels filter large quantities of water and can accumulate contaminants that are present in that water. The National Mussel Watch Program, of NOAA's National Status and Trends Program, periodically samples mussels at 12 Puget Sound stations and four along the outer Washington coast. NOAA scientists found in data from 1997 through 1998 that a few organic chemicals—especially PAHs—accumulated to higher concentrations in Puget Sound mussels than in mussels (and oysters) elsewhere around the United States coastline. In contrast, many metals (including arsenic, copper, lead, mercury and silver) were present at lower concentrations in Puget Sound mussels than observed in more remote areas including the outer coast.

PCBs have been of special concern in Puget Sound. During the 1997-98 National Mussel Watch Program total PCB concentrations in Puget Sound mussels ranged from 24 ppb (parts per billion or µg/kg), dry weight (dw), at Cape Flattery to 533

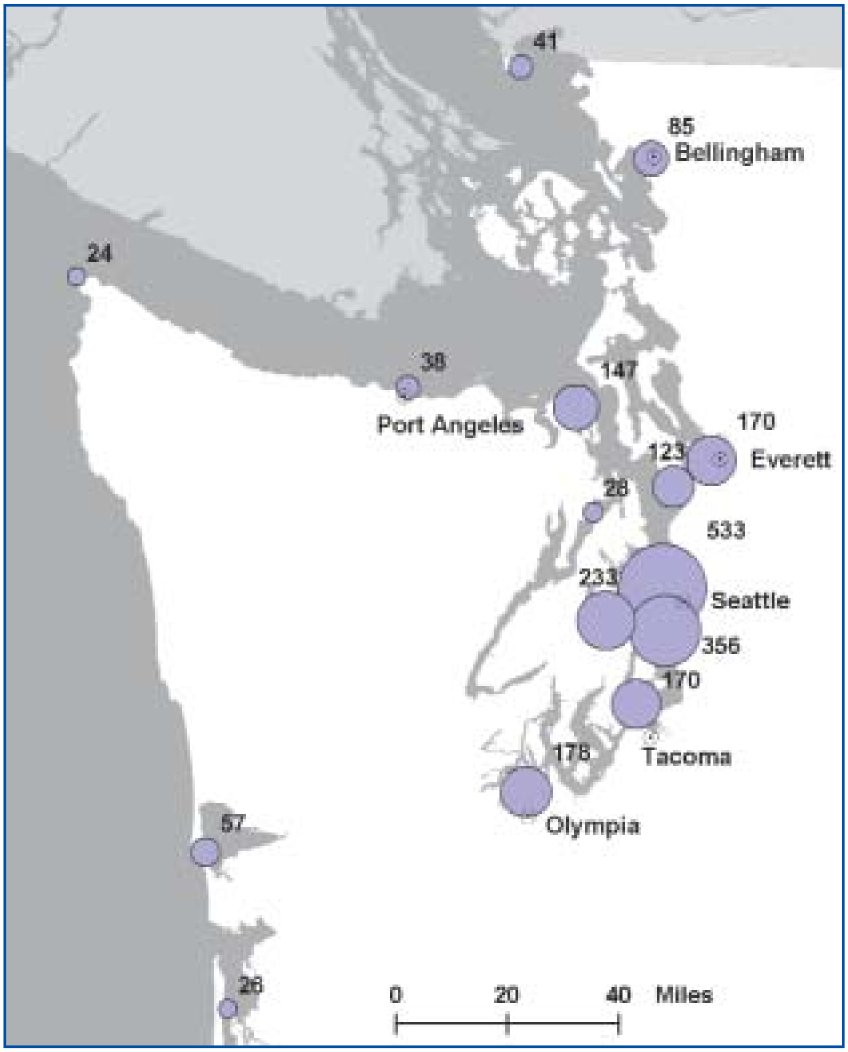
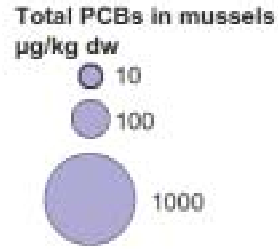


Figure 4-8. Total PCBs measured in Mussels, 1997-98.



Source: NOAA National Mussel Watch Program

ppb at Four Mile Rock beach near Magnolia (north Elliot Bay) with a median concentration of about 120 ppb dw and an average of 163 ppb dw (Figure 4-8). The Puget Sound median was slightly higher than the 1997-98 U.S. national median of 96 ppb dw but slightly lower than the national average of 255 ppb dw. Thus PCB concentrations in Puget Sound mussels were neither exceptionally high nor exceptionally low compared to the rest of the of the U.S.

NOAA scientists found that the outer Washington coast was not free of PCBs. In 1997 and 1998, PCB concentrations in mussels on the outer coast ranged from 24 ppb dw at Cape Flattery to 91 ppb dw at the Columbia River mouth. This information is important because it tells us that while concentrations in Puget Sound could be a lot lower, they won't be lower than what the coastal ocean is experiencing from other sources.

The NOAA National Mussel Watch data through 1998 provided evidence for multiple Puget Sound locations of long-term declining concentrations of the long-banned pesticides chlordane and DDT, and several metals such as lead and mercury. However, scientists also found that PCBs were no longer decreasing and were possibly increasing during the mid to late 1990s.

NOAA scientists now have PCB data from the 1999 and 2000 Mussel Watch surveys. The entire 16-year data record for selected stations in Puget Sound is shown in Figure

Figure 4-9. Time series of PCBs measured in mussels at selected sites in Puget Sound, 1985-2000.

□ **Port Townsend**

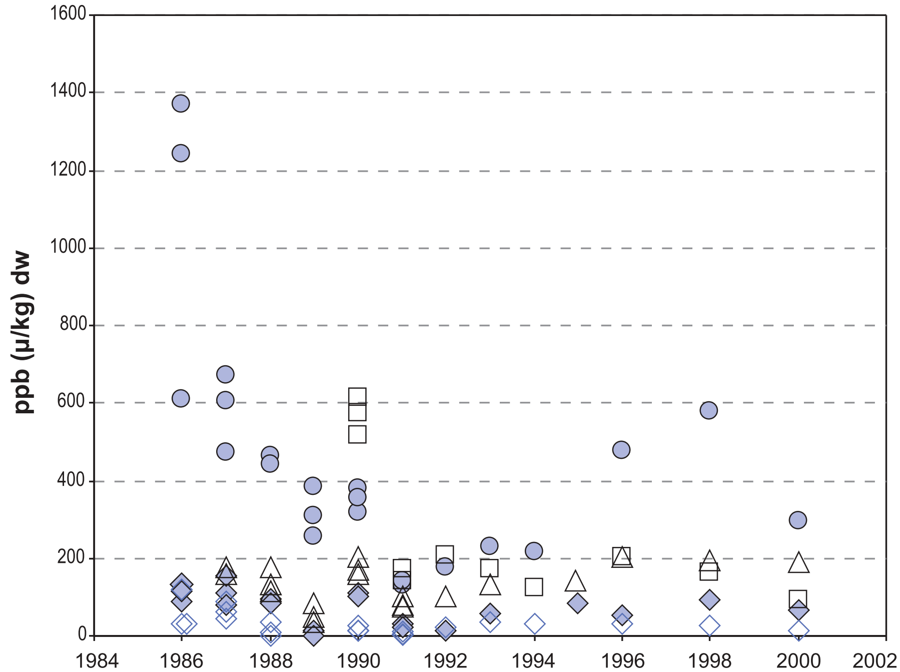
△ **Budd Inlet**

○ **Magnolia (4 Mile Rock)**

◆ **Squalicum Marina (Bellingham)**

◇ **Cape Flattery**

Source: NOAA National Mussel Watch Program



4-9. Three main features emerge from this time series. First, it is clear that concentrations of PCBs in mussels have generally declined during the past two decades. This decline is consistent with a Puget Sound decline that began in the 1960s and 1970s, as recorded in sediment layers in the bottom of Puget Sound. Second, regardless of the year, the highest concentrations were in mussels from central Puget Sound sites such as Four Mile Rock (Magnolia) and adjacent areas, consistent with this urban area as a long-term source. Third, the long-term downward trend was interrupted in the mid-1990s by increases at many locations. Fortunately, however, during 1999-2000, PCB concentrations in mussels began to decrease again.

We do not know the cause of this rise and fall during the past decade, but it occurred in remote areas (Washington coast, Alaska, etc.) as well as within Puget Sound. This suggests that mussels were experiencing PCB contamination caused or driven by some kind of large scale (ocean-wide) event or process. Possible causes include: increased atmospheric transport across the Pacific; increased input from land runoff; decreases in the food supply or growth rate of mussels (resulting in decreased tissue dilution) caused by prolonged periods of low plankton production associated with warm water conditions of the 1990s; or all of the above. Regardless of the cause, these data now indicate that we cannot be certain that PCBs will continue to decline at the dramatic rates seen during the 1970s to early 1990s.

Contaminants in Fish

Scientists at the state Department of Fish and Wildlife assess the status and spatial and temporal trends in contaminant accumulation in Puget Sound fishes and the effects of contamination on fish health. Currently they monitor English sole (*Pleuronectes vetulus*), demersal rockfish (*Sebastes* spp.), coho salmon (*Oncorhynchus kisutch*) and Pacific herring (*Clupea pallasi*). English sole have been monitored more than other species. This species has been sampled at 56 sites in Puget Sound and the Strait of Georgia, including eight core sites monitored annually. Since 1997, a substantial portion of this sampling effort has been redirected to better define small-scale spatial patterns (focus studies) in contaminant exposure and associated health effects for English sole and rockfish. Results from the focus studies on liver lesions in English sole from Elliott Bay (1997) and Sinclair Inlet (1998) were reported in the

2000 Update and the results of the Commencement Bay study (1999) are documented in this report (see Liver Disease in English sole). Currently, adult coho salmon returning to their natal streams are monitored biennially at five rivers in Puget Sound. In 1999, state Department of Fish and Wildlife scientists started monitoring contaminant exposure in adult herring stocks from northern, central and southern Puget Sound basins.

Pacific Herring

Following a successful pilot study of contaminant accumulation in individual herring from Fidalgo Bay in 1995 (described in the *2000 Update*), state Department of Fish and Wildlife scientists, in 1999, initiated an ongoing program to monitor contaminant levels in adult herring stocks from Puget Sound and the Georgia Basin. The objective was to measure contaminant exposure in adult herring from different spawning stocks from a broad geographic range to assess spatial variation in contaminant body burdens. Average contaminant exposure in adult spawning stocks should reflect environmental contamination from the geographic areas in which they reside. Furthermore, because herring are a short-lived species and only younger fish are sampled in this program, their contaminant loads reflect recent exposure to contaminants.

In 1999 state Department of Fish and Wildlife scientists sampled five spawning stocks (Figure 4-10): Denman/Hornby, Semiahmoo and Cherry Point, all in the Georgia Basin, Port Orchard in the central Puget Sound basin and Squaxin Pass in the southern Puget Sound basin. In 2000 they repeated their sampling on the Semiahmoo, Port Orchard and Squaxin Pass spawning stocks. Cherry Point fish spawn in April but all the other stocks complete their spawning by the end of February (Lemberg et al. 1997). Fish and Wildlife scientists estimated exposure to bio-accumulative organochlorines by measuring whole body concentrations of PCBs, chlorinated pesticides (DDT and its metabolites) and hexachlorobenzene (HCB). Recent exposure to PAHs, organic compounds that do not accumulate in fish, was estimated by concentrations of PAH metabolites, measured as Fluorescing Aromatic Compounds (FACs), in the fishes' bile.

Results from 1999 and 2000 indicate that Pacific herring from the central basin (Port Orchard), and to a lesser extent the southern Puget Sound basin (Squaxin Pass), had higher body burdens of PCBs than fish from more northern areas of Puget Sound and the Strait of Georgia (Table 4-2; O'Neill and West 2001). Lipid content of the samples also varied significantly among spawning stocks confounding the interpretation of these results. However, for the three stocks whose lipid values were similar to each other, (Denman/Hornby, Semiahmoo, and Port Orchard), PCB concentrations were highest in herring from the central Puget Sound (Figure 4-11). The elevated PCB concentration for Squaxin Pass fish, the most southern Puget Sound stock, was probably in part related to the significantly higher lipid levels observed for that spawning stock. However, the few Squaxin Pass samples with lipid levels that were similar to those from other locations had PCB concentrations that were more like those from Port Orchard than the Georgia Basin (Figure 4-11).

Significantly lower lipid levels were observed in the Cherry Point herring than all other stocks; however, for those individual samples where lipid levels were similar to those from fish in other Georgia Basin stocks, PCB concentrations were also similar. State Department of Fish and Wildlife scientists concluded that when differences in lipid content are taken into account (by expressing PCB concentration as per-lipid–weight basis), PCB concentrations in fish from central Puget Sound were significantly higher than at all other locations followed by fish sampled from Squaxin Pass and Cherry Point (Figure 4-12). PCB concentrations were lowest in fish sampled from the Strait of Georgia

Pacific Herring

Pacific herring are important prey to many other fish species, seabirds and marine mammals. Consequently, the health of these higher trophic levels is linked to the health of herring in the Puget Sound and Georgia Basin ecosystem. In the late winter and early spring, large spawning aggregations of adult herring provide rich sources of prey to piscivorous fishes and marine birds and mammals. Benthic fishes and seabirds consume the spawned eggs of Pacific herring. Larval and juvenile herring are also key components of the region's marine food web.

Because of their importance to the food web, there is concern that if herring accumulate toxic contaminants, much of the local food web could be affected.

Figure 4-10. Pacific herring sampling sites, 1995-2001.

Source: Washington State Department of Fish and Wildlife

Table 4-2. Mean (and standard deviations) organochlorine concentration (mg kg^{-1} wet weight) in Pacific herring stocks from the Georgia Basin and Puget Sound, 1999-2000.

Source: Washington State Department of Fish and Wildlife

	Mean Fish Age		% Lipids	Total PCBs		Total DDTs		HCB	
Stock	**(years)**	**N**	**Mean**	**Mean**	**s.d.**	**Mean**	**s.d.**	**Mean**	**s.d.**
Denman/Hornby	2.8	10	5.19	17.0	4.0	18.0	7.3	1.33	0.47
Cherry Point	2.8	10	3.59	54.9	13.0	12.7	3.0	0.35	0.19
Semiahmoo	2.8	20	5.62	52.3	24.7	17.8	7.0	1.54	0.27
Port Orchard	2.8	20	6.52	200.2	48.7	47.2	39.4	1.43	0.18
Squaxin Pass	2.9	19	9.15	165.8	48.8	20.9	5.0	1.43	0.27

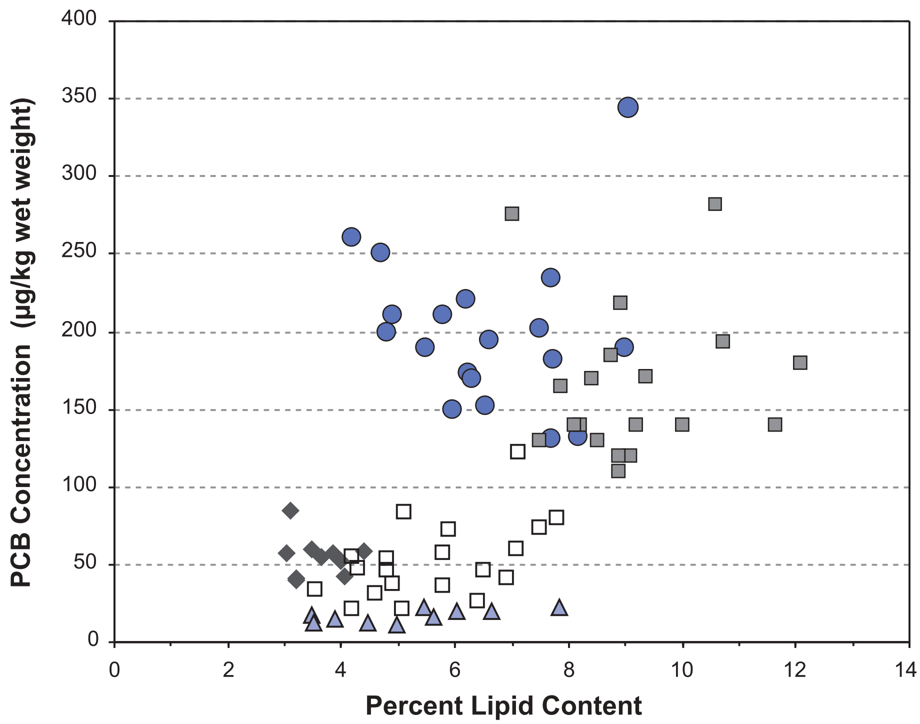


Figure 4-11. Relationship between lipid content and PCB concentration in whole bodies of Pacific herring from Georgia Basin and Puget Sound stocks, 1999-2000.

Source: Washington State Department of Fish and Wildlife

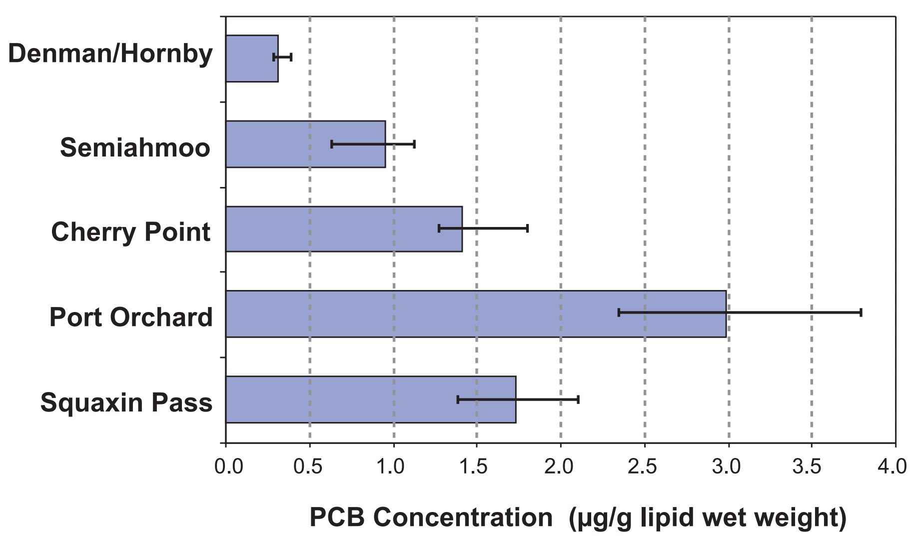


Figure 4-12 Median lipid-corrected PCB concentrations in whole bodies of Pacific herring from Georgia Basin and Puget Sound stocks, 1999-2000. Error bars indicate 25th and 75th percentile values.

Source: Washington State Department of Fish and Wildlife

State Department of Fish and Wildlife scientists also observed higher concentrations of DDTs (detected mostly as pp'-DDE, a metabolite of DDT) in fish from the Port Orchard stock (Table 4-2). The significance of these results is unclear as the concentration of pp'-DDE in Port Orchard fish was higher in 1999 than 2000. Low HCB concentrations were observed for all stocks but were lowest for the Cherry Point stock.

PAH Metabolites in Bile—Herring from the central Puget Sound also had higher PAH exposure than herring from other Puget Sound locations. Biliary FACs (PAH metabolites expressed as equivalents of benzo(a)pyrene and phenanthrene) were highest in herring from Port Orchard (Table 4-3). The Port Orchard bile samples also had significantly greater protein concentrations (a measure of bile diluteness that usually correlates positively with FAC concentrations) possibly explaining the elevated FAC levels. However, stock location was an important factor as well because for those

Table 4-3. Mean (and standard deviation) PAH metabolite concentrations measured in Pacific herring stocks from the Georgia Basin and Puget Sound, 2000. The metabolites include protein, benzo(a)pyrene (BaP) equivalents and phenanthrene (PHN) equivalents. The latter two are measured as fluorescing aromatic compounds (FAC).

Source: Washington State Department of Fish and Wildlife

		Protein (mg/ml)		FAC BaP (ng/g bile)		FAC PHN (ng/g bile)	
Stock	**N**	**Mean**	**s.d.**	**Mean**	**s.d.**	**Mean**	**s.d.**
Semiahmoo	10	6.62	2.63	224.6	68.5	22662	4074
Port Orchard	9	10.73	2.74	551.4	87.7	52237	4941
Squaxin Pass	10	5.77	2.86	305.0	91.4	26829	4955

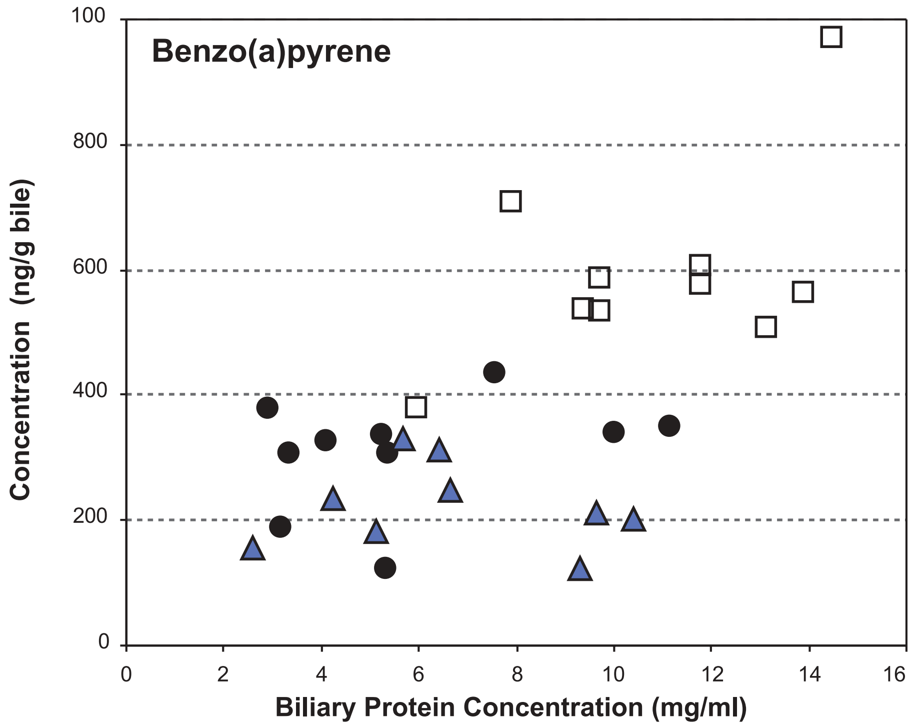
individual samples where protein values were similar among stocks, Port Orchard FAC levels were always higher than those observed in Semiahmoo and Squaxin Pass fish (Figure 4-13).

The higher toxics levels in herring from the central Puget Sound are likely associated with the greater sediment contamination, particularly in the industrialized bays and inlets, in this region of the Sound. Based on sediment sampling conducted by scientists from the Department of Ecology and NOAA (see previous **Sediment Contamination** section in this chapter), the central Puget Sound basin is known to have significantly higher concentrations of PCBs and PAHs, primarily from localized areas of contamination in urban bays, including Everett Harbor, Elliott Bay, Sinclair Inlet, Eagle Harbor, and Commencement Bay.

State Department of Fish and Wildlife scientists suggest that PCBs and PAHs are transferred from these contaminated sediments in the urban bays to the pelagic food web in the central basin and to a lesser extent the southern basin by variety of biological and physical processes. For example, macro-invertebrates and fishes associated with contaminated sediments may accumulate PCBs and PAHs that are then maternally transferred to their planktonic eggs and larvae, which in turn enter the herring's food web. Tidal action and storms may also re-suspend particulate matter containing PCBs and PAHs, that may then be transported to areas beyond the original contaminated sediment foot print. Re-suspension may also make PAHs and PCBs available to plankton and the pelagic food web of the central basin of Puget Sound and the relatively poorly flushed waters of south Puget Sound. In addition, new inputs continue from sources such as atmospheric deposition, surface water runoff and treated wastewater.

Through ongoing PSAMP studies, state Department of Fish and Wildlife scientists have documented that wild coho salmon from southern Puget Sound have higher lipid-specific PCB concentrations than fish from northern Puget Sound, possibly the result of their longer residence in central and southern Puget Sound during their out- and in-migrations (O'Neill et al. 1998). Similarly, scientists studying harbor seals have documented higher PCB concentrations in harbor seals in southern Puget Sound than in northern Georgia Strait (Calambokidis et al. 1988; Ross et al. 1998). Prey of these animals, such as Pacific herring, may be more contaminated in the more industrialized basins of central and southern Puget Sound than in the less developed basins of northern Puget Sound and Georgia Strait.

Based on a recent Adverse Effects Threshold for salmon developed by scientists with the National Marine Fisheries Service (Meador 2000), most herring from Puget Sound, with the exception of central Puget Sound fish, are not likely adversely affected by the levels of PCBs to which they are exposed. All of the Puget Sound herring PCB exposures measured to date from all sampling locations were well below the 50th-percentile threshold concentration (~12 µg/g lipid) that is associated with adverse effects in salmon. However, average PCB exposures in herring from the central Puget Sound basin were above the 10th-percentile concentration that is associated with adverse effects.



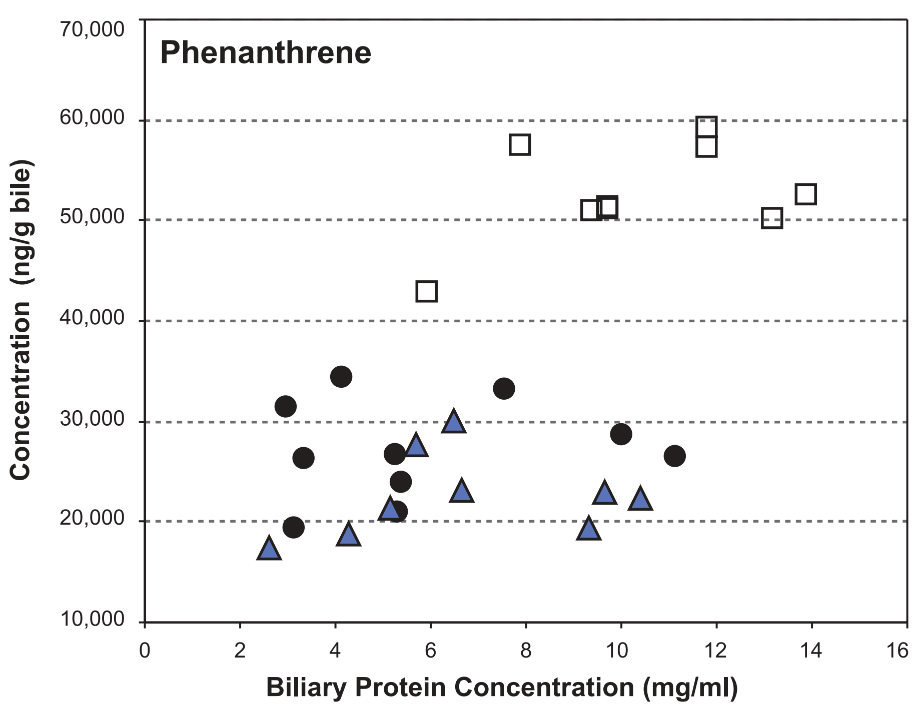


Figure 4-13. Relationship between concentration of biliary proteins and biliary FACs (estimated as equivalents of benzo(a)pyrene and phenanthrene) in Pacific herring stocks from the Georgia Basin and Puget Sound.

● **Squaxin Pass**

□ **Port Orchard**

▲ **Semiahmoo**

Source: Washington State Department of Fish and Wildlife

Future analyses should allow scientists to more accurately define spatial patterns in exposure of Pacific herring stocks in Puget Sound and the Georgia Basin to toxic contaminants. Ongoing monitoring should help to define the lipid:PCB, lipid:DDT, lipid:HCB and protein:bile FAC relationships, allowing better comparisons among sampling locations. State Department of Fish and Wildlife scientists have also initiated a study to assess whether organic contaminants are accumulated in spawned Pacific herring eggs.

Liver Disease in English Sole

Lesion prevalence in the livers of English sole is monitored as an indicator of contaminant-related fish health. Research by scientists with the National Marine Fisheries Service (NMFS) and the state Department of Fish and Wildlife has identified three primary risk factors associated with development of liver disease: (1) exposure to PAH contaminated sediments; (2) fish age; and (3) exposure to PCB contaminated sediments. Furthermore, reproductive impairment has also been observed in English sole at sampling sites with elevated occurrences of liver lesions (Johnson et al. 2001).

State and federal scientists continued their monitoring of the prevalence of liver disease at eight core sites in Puget Sound, including six sites that have been sampled regularly since 1989—non-urban sites in the Strait of Georgia and Hood Canal, a near-urban site in Port Gardner and three urban locations at Elliott Bay, Sinclair Inlet and Commencement Bay (Figure 4-14). Logistic regression analysis was used to calculate the risk of developing liver lesions in a population relative to a risk calculated for a reference population. The calculation of risk is based on the age distribution of the population as well as the liver lesion prevalence data. Scientists calculated risk at six core sites sampled annually between 1989 and 1999 against the baseline risk of developing liver lesions at 19 non-urban, relatively uncontaminated reference sites, sampled between 1989 and 1993. Relative risk of developing lesions at the reference site is 1.0 by definition. The calculation of relative risk from the prevalence data corrects for differences in fish age among sites.

Relative to baseline reference sites, the risk of developing liver disease at these core sites was highest at two urban bays (Elliott and Commencement), intermediate at Sinclair Inlet and Port Gardner, and lowest at non-urban sites in the Strait of Georgia and Hood Canal (Figure 4-15). This is one of the most powerful examples of impairment of fish health in Puget Sound that state and federal scientists have identified. Bottom dwelling English sole in two urban sites (Seattle waterfront in Elliott Bay and Thea Foss waterway in Commencement Bay), and one near-urban site (Port Gardner) had significantly higher risk of developing lesions than fish from reference areas in Puget Sound. PAH concentrations in sediments are also higher at these sites. At all other core sites, the risk of developing liver disease in English sole was not significantly different from reference areas in Puget Sound.

To further define the smaller spatial patterns in contaminant exposure and associated reductions in fish health, the state Department of Fish and Wildlife conducted focus studies in the three urban bays—Elliott Bay, Sinclair Inlet, and Commencement Bay. The team of scientists reported the results for liver disease in English sole for the Elliott Bay and Sinclair Inlet focus studies in the *2000 Puget Sound Update*. The results on liver disease in English sole sampled for the 1999 Commencement Bay focus study are presented in Figure 4-16.

Commencement Bay is a large urban embayment with many small waterways (or channels) at the head of the bay. Most of the industrial development is located in these waterways and the sediments there are significantly more contaminated than the

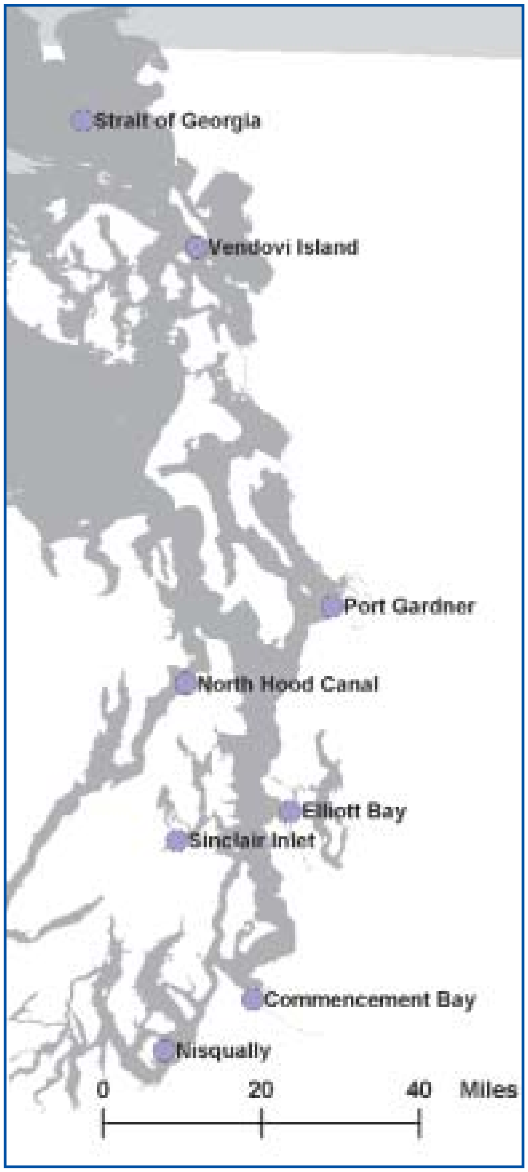


Figure 4-14. English sole sites sampled annually by PSAMP.

Source: Washington State Department of Fish and Wildlife

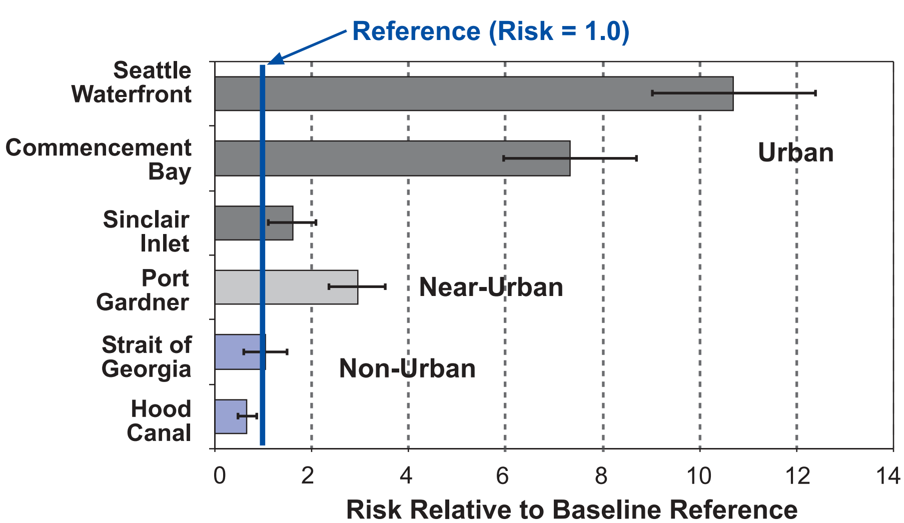


Figure 4-15. Average risk of developing liver disease in English sole at six core sites, sampled from 1989 through 2000, relative to risk at reference sites.

Source: Washington State Department of Fish and Wildlife

rest of the bay. Overall, the results from the 1999 focus study indicate that the risk of English sole developing liver disease in Commencement Bay is low and not significantly different than in baseline reference areas in Puget Sound (Figure 4-16). This focus study included the Thea Foss waterway that has been sampled by PSAMP since 1989.

Although the average risk of developing liver disease for English sole collected from the Thea Foss waterway in Commencement Bay between 1989 and 2000 was almost eight times higher than baseline reference sites in Puget Sound (Figure 4-15), risks in individual years were highly variable. In 1999, the risk was similar to baseline reference sites but in previous years it was as high as 16 times baseline reference (1993) and as low as 2.7 times baseline reference risk (1994). The high degree of variability observed in the risk of developing lesions in fish from the Thea Foss Waterway suggests that the sampled fish have not resided exclusively in the waterway during the several years over which liver lesions would have been developing.

Temporal Trends in Risk of Developing Liver Lesions in English Sole
Scientists with the state Department of Fish and Wildlife and NMFS have also monitored temporal trends in liver disease at these sites by estimating the risk of developing liver disease relative to baseline reference sites (non-urban sites sampled 1989 through 1993). The risk of liver disease increased in fish sampled along the Seattle Waterfront between 1989 and 1998 (see *2000 Puget Sound Update*), decreased in 1999 and was low again in 2000 (Figure 4-17). Scientists have not observed any trends in the risk of developing liver disease for English sole sampled from the other core sites.

State Department of Fish and Wildlife scientists reviewed the in-water activities that have taken place along the Seattle waterfront since 1989 that may have affected the risk of developing liver disease in English sole. Numerous sediment capping projects have been completed to the north, south and in the immediate vicinity of the Elliott Bay sampling site along the Seattle waterfront to sequester contaminated sediment. Capped sediment areas include: 2.8 acres in 1989 at the ferry terminal to the south; 3 acres in 1990 at the Denny Way Combined Sewer Overflow (CSO) to the north (Romberg et al. 1995); 4.5 acres in 1992 along Piers 53, 54 and 55, just inshore of the English sole sampling site, (Romberg et al. 1995); and 3.5 acres capped in 1994 at Bell Street Marina to the north.

Collectively, these projects should have lowered the PAH concentrations in surface sediments, reduced PAH exposure to English sole feeding in this area, and consistently lowered the risk of developing liver disease in these fish. There is evidence from other areas in Puget Sound that declines in liver disease in English sole associated with sediment capping projects are measurable with the type of sampling effort undertaken by PSAMP. Risk of developing liver lesions in English sole from Eagle Harbor declined significantly three to four years after the sediments in that area were capped with clean sediments and have remained consistently low (see next section).

The recent decline in risk of liver disease in Elliott Bay English sole suggests reduced exposure to PAHs; however, Elliott Bay sediment PAH concentrations actually increased during this period (Figure 4-18). The concentration of high molecular weight PAHs in the sediments at this site (monitored by King County) roughly doubled from 1993 to 2000. The King County monitoring station is located in 100 feet of water, and the end of Pier 63, between the Seattle Aquarium and the Bell Street Marina.

It is unknown if this observed increase in PAHs at the Seattle waterfront site is indicative of a broader increase of PAHs in Elliott Bay or whether this is a localized increase. Given the reductions in effluent associated with CSOs and the capping

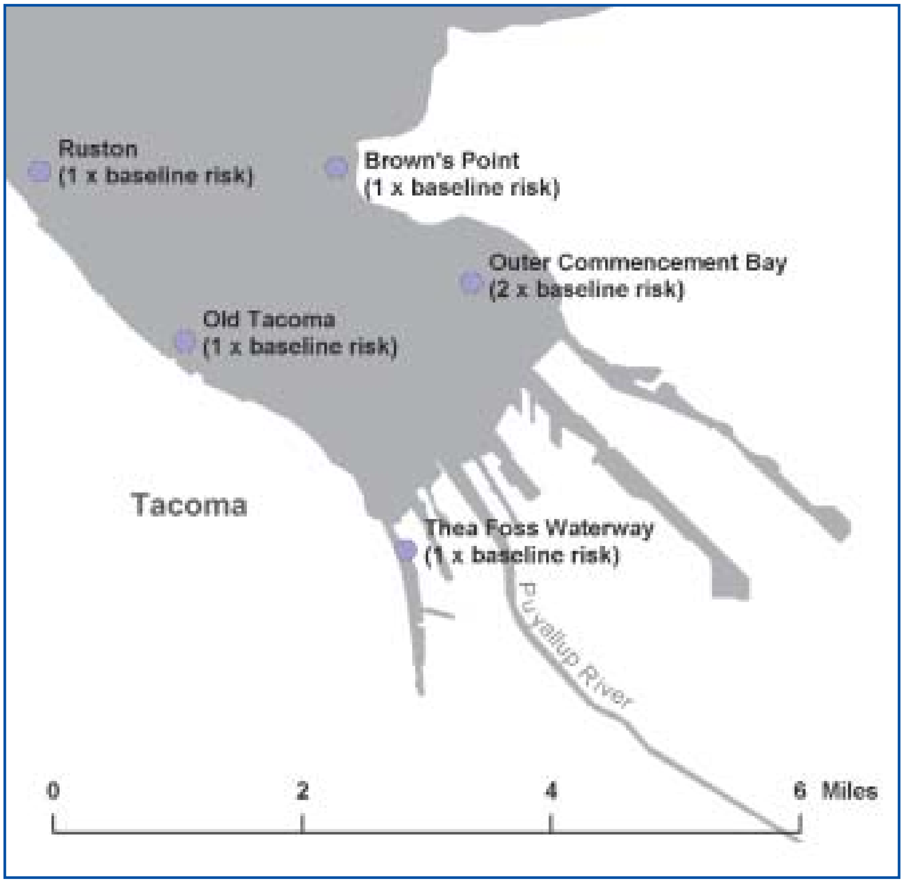


Figure 4-16 . Risk of developing liver lesions in English sole from various sites in Commencement Bay sampled in 1999, relative to baseline risk.

The risk at Thea Foss Waterway has been monitored since 1989 and is highly variable. While the figure reflects the relatively low risk seen at Thea Foss as part of the 1999 study, the average risk at this site from 1989 to 2000 was almost eight times higher than at the baseline reference sites.

Source: Washington State Department of Fish and Wildlife

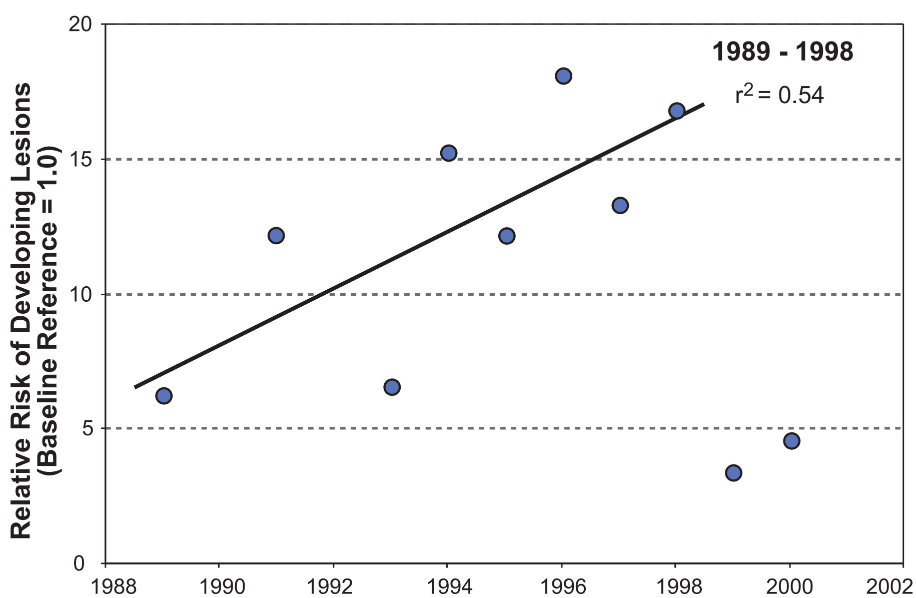
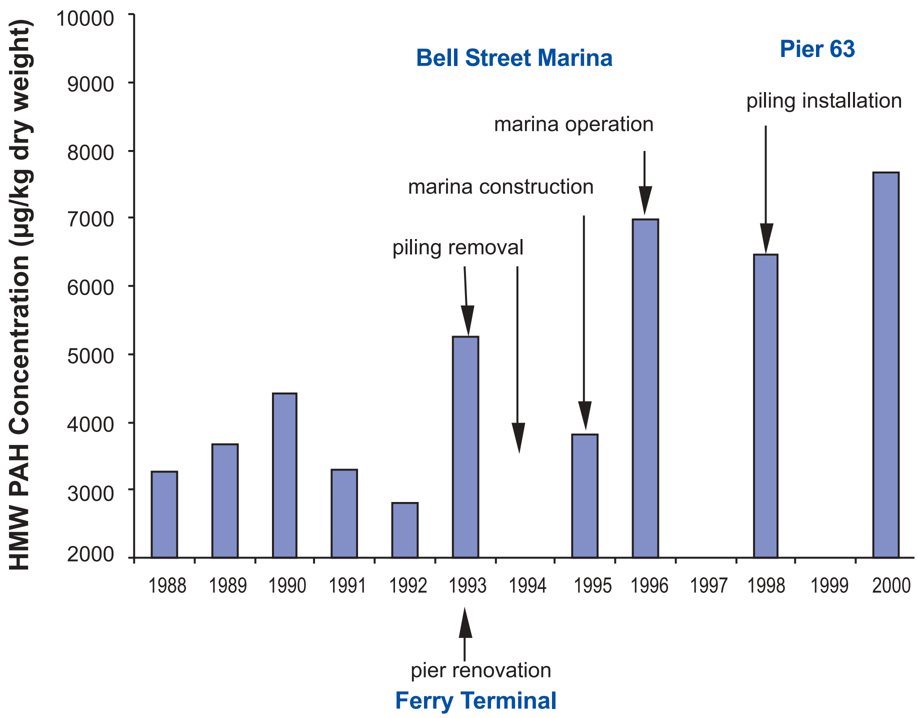


Figure 4-17. Risk of developing liver disease, relative to baseline reference, in English sole sampled along the Seattle Waterfront, 1989 – 2000.

Source: Washington State Department of Fish and Wildlife

Figure 4-18. High molecular weight PAH concentration in sediment along the Seattle Waterfront, 1989-2000, and the timing of major in-water activities.

Source: Washington State Department of Fish and Wildlife. Sediment data from King County Department of Natural Resources and Parks.



projects that have taken place along the Seattle waterfront, it is unlikely that there has been a bay-wide increase in the mass of PAHs in Elliott Bay. More likely, the concentration of PAHs at the Seattle waterfront site is a localized increase resulting from in-water activity associated with nearby pier modification (1993 to 1994 and 1998) and marina construction (1994 to 1995) (King County 1995). If this is the case, sediment PAH levels at the King County sediment site should decline in the future.

The increasing risk of English sole developing liver disease between 1989 and 1998 and the subsequent reduction 1999 and 2000 cannot be readily explained by the sediment chemistry at the King County monitoring station. From 1989 thru 1998, the risk of liver disease was well correlated with the average HPAH concentrations in sediments, but in 1999 and 2000, although the sediment PAHs remained high, the risk of developing liver disease declined (Figure 4-19).

To adequately interpret these results, state Department of Fish and Wildlife scientists would need additional information about the changes in PAH concentration along the whole of the Seattle waterfront and information on the home range of the English sole. Although the home range and feeding grounds of English sole is unknown, they likely extend over an area much greater than that represented by the sediments at the King County sampling site. Lacking such data, state Department of Fish and Wildlife scientists have several working hypotheses:

1. The 1999 and 2000 decline in the risk of developing liver disease may be due to an overall reduction to the mass of PAHs present in the surface sediments along the whole of the Seattle waterfront, independent of sediment changes observed at the specific site sampled over the years by King County.
2. The in-water activities described above may have displaced English sole, invalidating the assumption of a stable sampling population. Either direct physical disturbance during these activities, or less desirable habitat resulting from the placement of nonnative sediment may have led to this displacement.

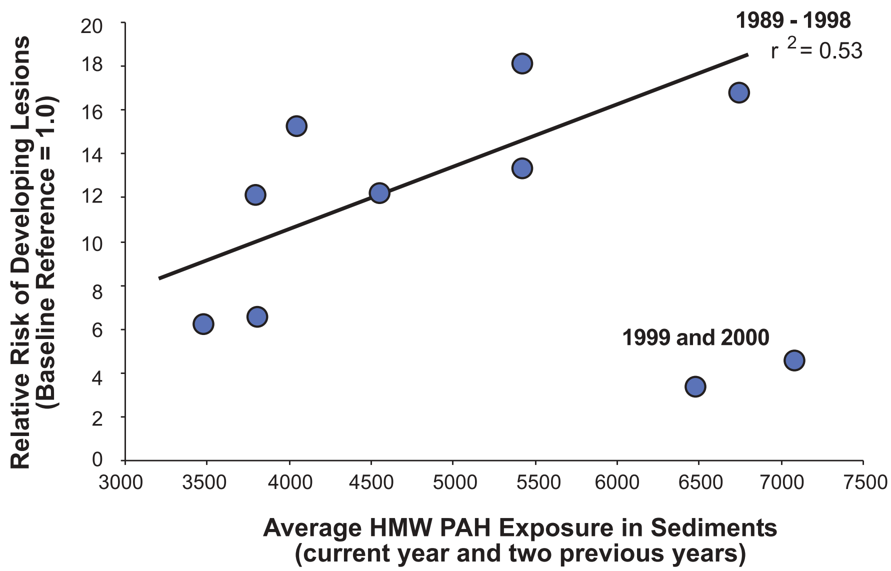


Figure 4-19. Relationship between the risk of developing liver disease in English sole, relative to baseline reference, and the average concentration of High Molecular Weight (HMW) PAHs in sediments.

Source: Washington State Department of Fish and Wildlife.

3. Finally, it is also possible that the observed increase in risk of liver disease was not a meaningful increase but rather a random variation in liver disease in English sole. State Department of Fish and Wildlife scientists plan to test this possibility with Monte Carlo simulations of risk of liver disease, to determine whether their sampling scheme is sufficient to overcome such variability.

Restoration of Fish Health Following Capping of Contaminated Sediment

In 1987, the U.S. Environmental Protection Agency identified Eagle Harbor, an embayment on Bainbridge Island in Puget Sound, as a Superfund site because of high sediment concentrations of PAHs released chronically from a nearby creosoting facility. Earlier studies at this site (1983-86) demonstrated high prevalences (up to approximately 75 percent) of toxicopathic (i.e. associated with exposure to chemical contaminants) liver lesions, including neoplasms (tumors), in resident English sole. NMFS scientists have demonstrated that neoplasia-related lesions can be induced experimentally by injections of a PAH-rich fraction extracted from Eagle Harbor sediment. Scientists have also studied the effects of PAH exposure in Eagle Harbor starry flounder and rock sole (1986-88) using several biochemical biomarkers, including hepatic CYP1A expression, biliary fluorescent aromatic compounds (FACs), and hydrophobic DNA adducts in liver. Prior to site remediation, hepatic lesion prevalences and biomarker values in these species from Eagle Harbor were among the highest found in Puget Sound.

The U.S. Environmental Protection Agency and Army Corps of Engineers placed a one-meter thick cap of relatively clean sediment (September 1993 to March 1994) over the most contaminated portions of Eagle Harbor in an attempt to sequester PAH-contaminated sediments. Scientists found that lesion prevalences and biomarker values just before capping began were generally reduced compared to historical data This is consistent with the creosoting facility closure and shore-based source controls.

Scientists found that toxicopathic liver lesion risk (a calculated parameter that considers lesion prevalence and fish age distribution), hepatic CYP1A, and biliary FACs from fish collected immediately after and at regular intervals up to two years after sediment capping, showed highly variable responses relative to values prior to cap placement. However, scientists found a significantly decreasing trend in risk for toxicopathic hepatic lesions in English sole (Figure 4-20) and rock sole, as well as for biliary FACs and hepatic DNA adducts in all three flatfish species, after approximately two years following the capping. Hepatic CYP1A levels showed no overall trend relative to time of sediment cap placement.

These results show that the sediment capping process has been relatively effective in reducing PAH exposure and associated biological effects in resident flatfish species, and that longer-term monitoring of pollutant responses in biological resources, such as resident fish, is critical to the demonstration of the efficacy of this type of sediment remediation.

Toxic Contaminants in Birds and Mammals

PCB Contamination in Orcas

Scientists from British Columbia (Ross et al. 2000) have reported "surprisingly high" concentrations of PCBs in the blubber of killer whales that reside in, or frequently visit, the coastal waters of British Columbia and Washington. The position of these whales atop a contaminated food web appears to be the primary reason for the observed levels of contamination.

The scientists analyzed blubber samples (biopsies), collected from orcas by dart between 1993 and 1996, for PCBs and chlorinated dioxins and furans. Samples were collected from the three major communities of orcas: transients, relatively elusive animals that occasionally visit the inland marine waters of British Columbia and Washington; southern residents, the orcas that spend much of the year in and around the southern Strait of Georgia and Puget Sound; and northern residents, a group of orcas that reside primarily in protected marine waters from the northern Strait of Georgia to southeast Alaska. The transient orcas, which feed primarily on seals, sea lions and other marine mammals, are among the most highly contaminated marine mammals in the world with total PCB concentrations of 250 milligrams per kilogram of lipid (mg/kg lipid) in males and 59 mg/kg lipid in females.

Resident orcas feed lower on the food web, primarily on fish and especially salmon, and their blubber contains lower PCB concentrations than observed in the transient whales. Males from the southern resident community showed the highest PCB contamination observed among residents, 150 mg/kg lipid. Based on extrapolations from their data, the scientists estimate that the southern resident orcas are four to six times more contaminated than the northern residents, presumably because their prey include more highly contaminated fish from the industrialized parts of Puget Sound and the southern Strait of Georgia.

Contaminants in Bald Eagles Nesting in Hood Canal

A recently completed study investigated levels of toxic contaminants in eggs, blood and prey of bald eagles (*Haliaeetus leucocephalus*) nesting in the Hood Canal area (Mahaffy et al. 2001). Another component of this study tracked the reproductive success of the Hood Canal bald eagle population (Chapter 6). The bald eagle is currently listed as a threatened species under the Endangered Species Act but is proposed for removal from the List of Endangered and Threatened Wildlife (USDI 1999). At the time the species was listed, environmental contaminants such as DDT were cited as the primary reason for its decline. Currently, bald eagle numbers are increasing in Washington State, but the U.S. Fish and Wildlife Service initiated this study because of the depressed bald eagle productivity in Hood Canal relative to Washington State as a whole.

U.S. Fish and Wildlife scientists collected 13 bald eagle eggs from Hood Canal territories in 1992 through 1993 and 1994 through 1997 and five additional eggs

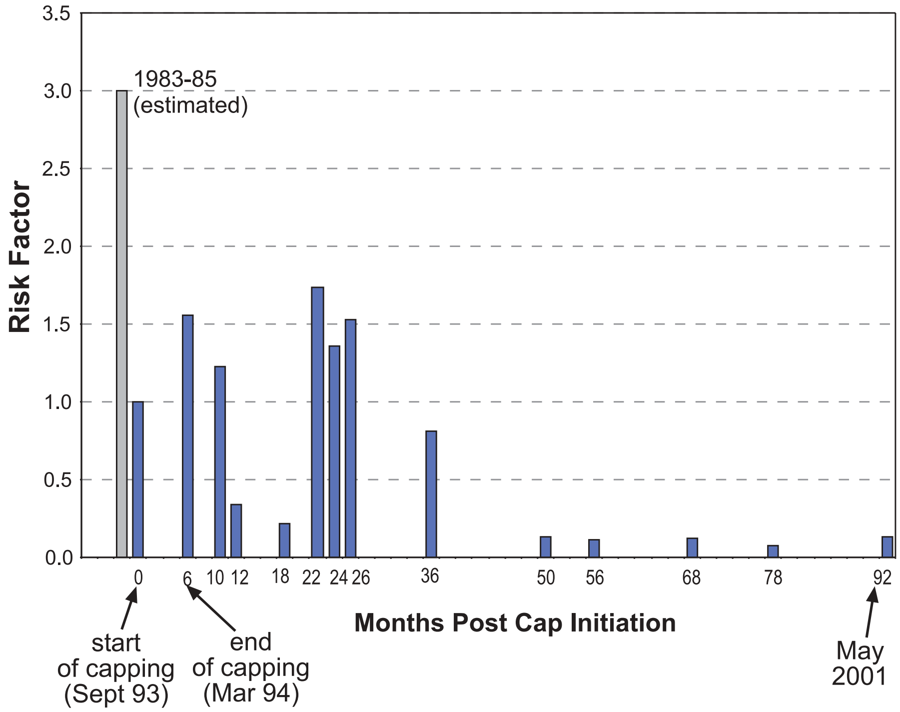


Figure 4-20. Risk of developing toxicopathic liver lesions in English sole from Eagle Harbor, prior to and following capping of contaminated sediment at this site, relative to risk at the time of capping.

Source: National Marine Fisheries Service

from territories outside Hood Canal. In addition, they sampled blood from the Hood Canal birds, and prey fish and sediments from Hood Canal for the measurement of contaminants.

If environmental contaminants were the cause of low productivity, then nests with the highest failure rates would be expected to have eggs with the highest contaminant concentrations. Twelve of the 13 Hood Canal eggs exceeded the 4.0 µg/g threshold total PCB level estimated for normal reproduction of bald eagles. Only one egg collected outside Hood Canal exceeded the 4.0 µg/g total PCB level. Based on these results, Mahaffy et al. (2001) conclude that PCBs likely reduced productivity in the Hood Canal bald eagle territories. In contrast, concentrations of DDE, a breakdown product of DDT, were less than concentrations considered to have an impact on bald eagle productivity.

Concentrations of mercury, selenium and arsenic were below levels of concern in the five eggs where they were analyzed. Detectable levels of PCBs and p,p'-DDE were present in blood samples. Contaminants measured in the fish and sediment samples from Hood Canal were not great enough to account for the elevated amounts found in bald eagles from this area. These results suggest that the contaminant pathway to bald eagles in Hood Canal is likely from food items other than fish, such as birds or marine mammals.

Acting on the Findings

The results presented in this chapter suggest that while we are still trying to understand the extent and effects of toxic contamination in ecosystems, we can successfully manage contamination in some specific cases. The reduction in risk of developing liver lesions in English sole following the capping of contaminated sediment at Eagle Harbor is an excellent example. Continued monitoring is necessary to evaluate such management actions. The need for continuing monitoring is particularly compelling when viewed in the light of new results that reveal toxic effects in organisms where they were previously unknown and studies that identify new compounds as toxic threats.

Some specific recommendations follow directly from the results of studies presented in this chapter:

- As much as possible, studies should be interdisciplinary in nature such that contaminant data can be integrated with population data and life history patterns. Understanding of the currently unexplained variability in some contaminant data will require such an approach.
- Further pilot studies are needed to assess toxic contaminant impacts in previously unstudied species to fully evaluate ecosystem effects of these contaminants.
- Continued monitoring is needed for biota affected by contaminants, even when contaminant levels and productivity are improving, as long as a contaminant impact is observed. This will ensure that recovery proceeds as expected, and important causal factors have not been overlooked.
- Further studies are needed to better understand sources of the recent increases in benzoic acid seen in sediments and shellfish as well as the ecological implications of these increases.
- For contaminants that are increasing in Puget Sound sediments, scientists needs to quantify sources and policy makers need to determine if current controls are inadequate to control these pollutants.
- The state Department of Fish and Wildlife needs to further investigate the source and pathways of contaminants in herring. Emphasis should be on assessing whether (1) dredged material management; (2) contaminated sediment cleanup; or (3) wastewater discharge control could reduce herring contaminant exposure.
- Scientists need to use developing food web models for Strait of Georgia and south Puget Sound and information on contaminant burdens in various organisms to see if they can accurately describe the major pathways of accumulation to rockfish, salmon, harbor seals, orcas and Hood Canal eagles. These studies can identify gaps and encourage additional study to fill these gaps. This work can also identify the leading opportunities to reduce accumulation of toxics in the food web.
- Scientists need to keep up efforts to relate fish contamination and disease to sediment contamination, especially at areas such as Seattle Waterfront and Thea Foss Waterway to try to learn more about how fish respond to cleanup efforts and sediment disturbances.
- More focused monitoring is needed to measure the effectiveness of alternative contaminant control measures.